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PROCEEDINGS  
OF THE  
**NATIONAL ACADEMY OF SCIENCES, INDIA**  
**2001**

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VOL. LXXI

SECTION-A

PART IV

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## **Determination of sulphide sulphur in alkaline solutions**

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Received July 24, 2000; Revised February 6, 2001; Accepted May 9, 2001

### **Abstract**

Conditions have been developed for the indirect titrimetric determination of water soluble sulphide sulphur with ferricyanide as a reagent using copper (II) sulphate as catalyst. The excess ferricyanide content is titrated with standard hydroxylamine solution using tris (4,7-dihydroxy-1,10-phenanthroline) – Iron (III) as an indicator in strong alkali medium. The proposed method for the determination of sulphide sulphur is simple, rapid, and sensitive (0.3 to 20 mg). The pooled standard deviation and average error are found to be 0.04 mg and  $\pm 0.44\%$  respectively.

(Keywords : titrimetry/sulphide sulphur/ferricyanide)

### **Introduction**

The development of fairly effective analytical techniques becomes significant towards control methodologies in pollution studies. Sulphide sulphur is one such pollutant. As such a procedure that can be used to determine sulphide content with a reasonable degree of accuracy in the presence of some other sulphur compounds will have its own definite advantages. The various titrimetric methods reported for determining sulphide sulphur in different materials and waster waters were generally based on the reagents like iodine<sup>1-4</sup>, permanganate<sup>5-6</sup> and others<sup>7-10</sup>.

Most of these methods are usually associated with one or other drawbacks and limitations; for example, instability in acid solution, interference due to other sulphur compounds, requiring potentiometric assembly, high temperatures, which have been well

exemplified<sup>11-12</sup>. The reported methods<sup>12-14</sup> for the determination of sulphide sulphur with ferricyanide under controlled pH in the absence of oxygen depend on its oxidation to elemental sulphur. Amperometric method reported needs the use of osmium tetroxide as catalyst and temperature while potentiometric methods have necessarily to be carried out at elevated temperatures. But so far no room temperature titrimetric visual end-point method could be found in the literature for the determination of sulphide sulphur based on its oxidation to the sulphate state with ferricyanide.

The present communication describes a rapid and convenient back titration method to the determination of sulphide using ferricyanide as oxidant. The procedure is based<sup>15</sup> on the titration of excess ferricyanide content by hydroxylamine using tris (4, 7, dihydroxy-1, 10-phenanthroline)-Iron(III) [DHP]<sub>3</sub> – Iron(III)] as an indicator.

### Materials and Method

All chemicals used were of analytical grade reagents. 0.1N solutions of potassium ferricyanide and hydroxylamine hydrochloride were prepared and standardized<sup>16</sup>. 0.25% copper (II) sulphate solution was prepared. 0.16% (DHP)<sub>3</sub> – Iron(III) indicator solution

Table 1 – Effect of Copper (II) Sulphate.  
Taken sulphide = 5.44 mg

Time of reaction (min.)	Sulphide found (mg)		% Recovery of sulphide	
	In the absence of Cu(II)	$1.7 \times 10^{-4}$ M Cu(II)	In the absence of Cu (II)	In the presence of $1.7 \times 10^{-4}$ M Cu(II)
1	3.64	5.20	66.9	96.0
2	3.84	5.31	70.6	97.6
3	4.08	5.43	75.0	99.8
4	4.34	5.44	79.8	100.0
6	4.84	5.39	88.9	99.1
8	5.18	5.41	92.9	99.4
10	5.38	5.38	98.9	98.9
20	5.44	5.42	100.00	99.6
30	5.42	5.44	96.6	100.0

Table 2 – Determination of sulphide sulphur

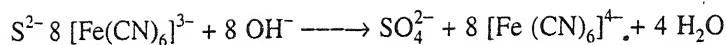
Sulphide Sulphur (mg)			
Taken	Found		
	In pure samples	In the presence of 1.42mg sulphite	In the presence of 20.82 mg of thiosulphate
0.450	0.454	0.456	0.454
0.912	0.916	0.918	0.916
2.736	2.726	2.728	2.730
4.560	4.548	4.544	4.542
9.120	9.149	9.142	91.53

was prepared as reported<sup>17</sup>. Solutions of 0.01 M sodium sulphide ( $\text{Na}_2\text{S} \cdot 9\text{H}_2\text{O}$ ), 0.05M sodium sulphite and 0.05M sodium thiosulphate were prepared and standardized<sup>12</sup>. These solutions were prepared on the day of use. A 6N sodium hydroxide solution was prepared.

Take an aliquot of sulphide (0.3 to 20mg) and 30 ml of 6N sodium hydroxide into a conical flask. Add a known excess volume of ferricyanide and then 1ml of 0.25% copper (II) solution to it. Keep the reaction mixture aside for 3 min. Add 0.5ml of  $(\text{DHP})_3$  – Iron (III) indicator solution. At this stage the colour of solution is greenish yellow. Titrate the excess ferricyanide content with standard hydroxylamine solution until the colour changes from greenish yellow to intense red. A blank titration on the same ferricyanide volume is run.

### Results and Discussion

Preliminary experiments in the determination of sulphide sulphur with ferricyanide revealed that the minimum time required for the complete oxidation of sulphide to sulphate is 10 min. The reaction time gets reduced to 3 min. in the presence of copper (II) as catalyst or temperature (50-60°C). The  $[\text{Ferricyanide}]/[\text{Sulphide}]$  should be greater than 89 to give results corresponding to the indicated accuracy. The order of addition of reagents should be strictly followed as given in the procedure. The results in Table 1 clearly show the positive catalyst effect of copper (II) on the rate of reaction between sulphide and ferricyanide. The stoichiometry of the reaction can be expressed by the following equation.



The addition of formaldehyde eliminates the interference of sulphite which forms an inert complex. 5-fold excess of sulphite and 100-fold excess of thiosulphate do not interfere with the determination.

The results of the proposed method for the determination of sulphide sulphur in synthetic mixtures are in good agreement with the standard method. The pooled standard deviation and average error of the present method are found to be 0.04mg and  $\pm 0.44\%$  respectively. The results in the determination of sulphide are given in Table 2. Some of the advantages of the present method are :

- I. Does not require close control of pH.
- II. Thiosulphate and sulphite do not interfere
- III. Use of cheaper catalyst [Cu(II) as compared to Os (VIII)]

The method is more conveniently suitable for the estimation of sulphide sulphur in refinery spent caustic solutions as compared to the method described<sup>18</sup>.

#### Acknowledgements

This work belongs to a research project, funded by UGC, New Delhi (India). The author thanks Prof. Dr. U. Muralikrishna for his guidance and Miss. T.A.S.S. Santhisree for checking some of the results reported in this paper.

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## Studies on oxovanadium (IV) and dioxouranium (VI) complexes with pyrazine-2-and pyrazine-2, 3-carboxylic acid hydrazide

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Received February 12, 2000; Final Revision March 16, 2001; Accepted May 31, 2001

### Abstract

Complexes of oxovanadium (IV) and dioxouranium (VI) with pyrazine-2-carboxylic acid hydrazide (L) and pyrazine-2, 3-dicarboxylic acid dihydrazide (L') have been prepared and characterized through analytical, spectral and magnetic moment data.  $\text{VO}(\text{acac})_2$  with L gives oxovanadium (IV) complex of the Schiff's base generated *in situ* from the reaction of the hydrazide and 'acac' moiety; with L',  $\text{VO}(\text{IV})$  and  $\text{UO}_2(\text{VI})$  result in isolation of complexes of pyrazine-3-hydrazide-2-carboxylic acid presumably derived *in situ* from the hydrolysis of a hydrazide group.

(Keywords : metal complexes/pyrazine carboxylic acid hydrazides/ $\text{VO}(\text{IV})/\text{UO}_2(\text{VI})$ /Schiff's base)

### Introduction

Metal complexes of carboxylic acid hydrazides are interesting due to their biological and structural importance<sup>1-3</sup>. Our previous investigation<sup>4,5</sup> using pyrazine-2-carboxylic acid hydrazide (L) and pyrazine-2, 3-dicarboxylic acid dihydrazide (L') with several centres of coordination have prompted studies on their oxovanadium (IV) and dioxouranium (VI) halides, sulphates, acetates and acetylacetones.

Vanadium is receiving considerable attention as a biologically important metal due to the existence of vanadoenzyme<sup>6</sup>. Dioxouranium (VI) complexes are of structural interest due to varied geometry they assume, of late dioxouranium (VI) complexes are said<sup>7</sup> to have possible applications in solar energy conversion systems owing to spectral and excited state electron transfer properties of the  $\text{UO}_2^{2+}$  ion.

### Materials and Method

Pyrazine-2-carboxylic acid hydrazide (L) and pyrazine-2, 3-dicarboxylic acid dihydrazide (L') (Fig. 1 & 2);  $\text{VO}(\text{acac})_2$  and  $\text{UO}_2(\text{acac})_2 \cdot 2\text{H}_2\text{O}$ ; and  $\text{VOCl}_2$  (ethanolic

solution, pH ~ 3-4) were prepared following published procedures<sup>4,5,8,9,10</sup>. VOSO<sub>4</sub>.H<sub>2</sub>O and UO<sub>2</sub>(CH<sub>3</sub>COO)<sub>2</sub>.2H<sub>2</sub>O used were E Merck/BDH quality.

Spectral (IR, UV-Vis, NMR, EPR), magnetic moments and conductivity measurements and C, H, N analyses were done as described elsewhere<sup>4,5</sup>. Metal, halogen and sulphur were analysed gravimetrically by standard methods as oxides (V<sub>2</sub>O<sub>5</sub>, U<sub>3</sub>O<sub>8</sub>) and as AgCl and BaSO<sub>4</sub>. Molecular weights were determined by Rast method with diphenyl as solvent. FAB-MS using Jeol SX-102 instrument were recorded in CDRI, Lucknow.

*Complex 1* : V<sub>2</sub>O<sub>5</sub> (0.470 g, ~2.58 mmol) was refluxed on a steam bath with dry ethanol (10 ml) containing 12 M HCl (5 ml). A clear green solution of VOCl<sub>2</sub> thus obtained was filtered in a basin and the filtrate concentrated to ~1 ml. The resultant solution (pH 3-4) after dilution with dry ethanol (5 ml) was mixed with solution of ligand L (145 g, ~10.51 mmol) in hot ethanol (50 ml). Immediate colour-change occurred and a red compound appeared. The suspension was refluxed (2h) on a steam bath. The red compound was filtered hot, washed with dry ethanol and dried over fused CaCl<sub>2</sub> in a desiccator. Yield : 60% m.p. 270°C (dec.).

*Analy. data* : found % (calc for VOLCl<sub>2</sub>.4H<sub>2</sub>O) M 14.04 (14.65), C 16.57 (17.24), H 4.17 (4.02), N 15.88 (16.09), Cl 18.60 (20.46).  $\Lambda_M$  ( $\Omega^{-1} \text{cm}^2 \text{mol}^{-1}$ , DMSO) : 146.1.  $\mu_{\text{eff}}$  (RT) : 1.71 BM. EPR (RT) :  $g_{\text{av}} = 1.97$ . IR (KBr),  $\text{cm}^{-1}$  : 3000-3500 (br), 1620(s), 1040 (ms), 970(s), 1240 (wsh), 1170-1140 (s), 1050-1030 (s); and UV-Vis (nujol-mull),  $\text{cm}^{-1}$  : 22730 (br), 19230 (br, sh), 13330 (uw).

*Complex 2* : A solution containing VOSO<sub>4</sub>.H<sub>2</sub>O (0.900 g, 5 mmol) in dry methanol (50 ml) was mixed with solution of ligand L (1.65 g, ~ 12 mmol) in the same solvent (50 ml) and the mixture was refluxed (4h) on a steam bath when a blackish compound formed was filtered hot and washed with hot methanol. The compound was purified by dissolving in hot water, filtering and evaporating the filtrate to dryness to obtain brown crystals which were dried in a desiccator over fused CaCl<sub>2</sub>. Yield 56%, m.p. >300°C.

*Analy. data* : found % (calc. for VOLSO<sub>4</sub>.2H<sub>2</sub>O) M 14.81 (15.13), C19.13 (17.80), H 3.15 (2.97), N 16.75 (16.62), SO<sub>4</sub> 28.86 (28.48).  $\Lambda_M$  ( $\Omega^{-1} \text{cm}^2 \text{mol}^{-1}$ , DMSO) : 62.3.  $\mu_{\text{eff}}$  (RT) : 1.69 BM. EPR (RT) :  $g_{\text{av}} = 1.96$  IR (KBr),  $\text{cm}^{-1}$  : 3000-3500 (br), 1620 (s), 1030 (m); and UV-Vis (nujol-mull) 23810 (br)  $\text{cm}^{-1}$ .

*Complex 3* : A mixed solution containing VO(acac)<sub>2</sub> (0.160 g, ~0.60 mmol) in dry methanol (10 ml) and solution of ligand L (0.165 g, ~ 1, 2 mmol) in the same solvent (10 ml) was refluxed (2h) on a steam bath. Brown coloured crystalline product separated was filtered hot, washed with hot methanol and dried over fused CaCl<sub>2</sub> in a desiccator. Yield 60%, m.p. 240°C (dec.)

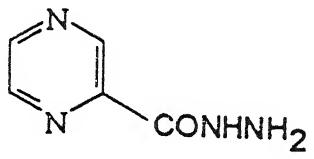


Fig. 1. (L)

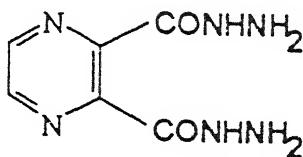


Fig. 2. (L')

*Analy. data* : found % (calc. for  $(VO)_2(acacL)_2$ ) M 18.82 (17.89), C 41.34 (42.10), H 3.55 (3.51), N 19.63 (19.65). FAB-MS molecular peak observed at M/Z 570.  $\Lambda_M$  ( $\Omega^{-1} \text{ cm}^2 \text{ mol}^{-1}$ , DMSO) : 46.6  $\mu_{\text{eff}}$  (RT) : 1.55 BM/V. EPR (RT) :  $g_{\text{av}} = 1.96$  IR (KBr),  $\text{cm}^{-1}$  : 1580 (s), 1420 (m), 530 (s), 840 (m, sh) 970 (s); and UV-Vis (nujol-mull),  $\text{cm}^{-1}$  : 22730 (br), 17860 (sh), 16670 (sh), 15150 (sh). \*acacL = Schiff base (dianionic) from L and acac ion.

*Complex 4* : A mixed solution containing  $UO_2(acac)_2 \cdot 2H_2O$  (0.500 g, ~.1 mmol) in dry methanol (20 ml) and ligand L (0.280 g, ~2.03 mmol) in the same solvent (20 ml) was refluxed (2h) on a steam bath. the precipitated brown complex was filtered hot washed with hot dry methanol and dried over fused  $CaCl_2$  in a desiccator. Yield : 45%, m.p. >300°C.

*Analy. data* : found % (calc. for  $UO_2(L-H)_2$ ) M 44.50 (43.75), C 22.25 (22.05), H 2.09 (1.84), N 21.55 (20.59).  $\Lambda_M$  ( $\Omega^{-1} \text{ cm}^2 \text{ mol}^{-1}$ , DMSO) : 26.0. IR (KBr),  $\text{cm}^{-1}$  : 1600(s), 1040 (ms), 12.80 (ms), 840 (s), 920 (s); UV-Vis (nujol-mull),  $\text{cm}^{-1}$  23260 (br), 16400 (sh);  $^1H$  NMR,  $\delta$  (ppm) : 5.08 (NH<sub>2</sub>) (Fig. 3).

*Complex 5* : To a solution (1 ml) of  $VOCl_2$  prepared from  $V_2O_5$  (0.960 g, 5.18 mmol) as a complex 1 and diluted with dry methanol (10 ml) was added to suspension of ligand L' (1.01 g, ~5.15 mmol) in dry methanol (100 ml) and the whole refluxed (3h). The suspension became clear and was filtered. The filtrate was concentrated (1/2 v), refrigerated and then mixed with a little ether. Brown complex formed was filtered, washed with cold methanol and dried over fused  $CaCl_2$  in a desiccator. Yield 40%, m.p. 290°C (dec.).

*Analy. data* : found % (calc. for  $(VO)_2L' (OCH_3)_2Cl_2 \cdot 2H_2O$ ) M 19.13 (20.44), C 21.55 (19.24), H 3.84 (3.61), N 16.97 (16.83), Cl 15.90 (14.23).  $\Lambda_M$  ( $\Omega^{-1} \text{ cm}^2 \text{ mol}^{-1}$ , DMSO) : 97.1.  $\mu_{\text{eff}}$  (RT) : 1.70 BM/V. EPR (RT) :  $g_{\text{av}} = 197$ . IR (KBr),  $\text{cm}^{-1}$  : 2500–3500 (br), 1650

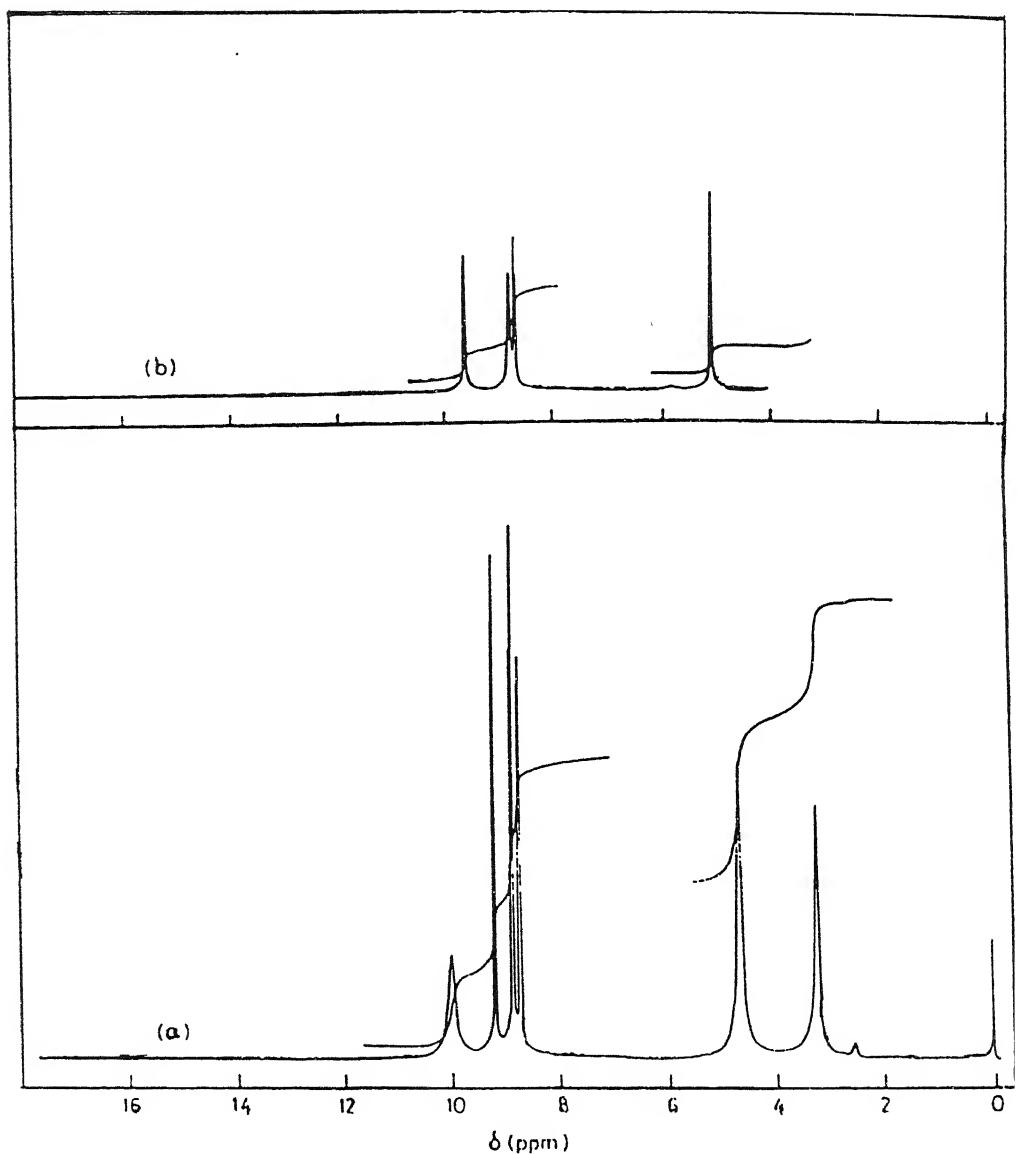


Fig. 3 – <sup>1</sup>HNMR spectra of (a) L and (b) complex 4 in DMSO-d<sub>6</sub> (Ref. TMS)

(intensity largely reduced), 450-460 (ms); and UV-Vis (nujol-mull),  $\text{cm}^{-1}$  : 25000 (br), 13890 (vw).

*Complex 6* : A solution of  $\text{VO}(\text{acac})_2$  (1.2 g, ~4.53 mmol) in dry methanol (50 ml) was mixed with a suspension of ligand  $\text{L}'$  (0.900 g ~4.60 mmol) in dry methanol (100 ml) and the whole refluxed (5h) on a steam bath when the suspension became clear. The solution was filtered and the filtrate was concentrated (1/2 v), refrigerated and then mixed with a little ether. Ash coloured complex that settled down was filtered, washed with chilled methanol and dried over fused  $\text{CaCl}_2$  in a desiccator. Yield : 38% m.p. > 300°C.

*Analy. data* : (calc. for  $(\text{VO})_2(\text{OH})_2(\text{Pz-3-CONHNH}_2\text{-2-COO})_2$ ) M 20.00 (19.24), C 27.67 (27.17), H 2.71 (2.26), N 20.24 (21.13).  $\Lambda_M$  ( $\Omega^{-1} \text{cm}^2 \text{mol}^{-1}$ , DMSO) : 31.9  $\mu_{\text{eff}}$  (RT) : 1.17 BM/V EPR (RT) :  $g_{\text{av}} = 1.97$  IR (KBr),  $\text{cm}^{-1}$  : 1650 (s), 1590 (ms), 1330 (ms), 965 (s), 460 (s), 580 (m); and UV-Vis (nujol-mull), 25000 (br)  $\text{cm}^{-1}$ .

*Complex 7* : A solution of  $\text{VOSO}_4\text{H}_2\text{O}$  (0.180 g, ~1.10 mmol) in dry methanol (20 ml) was mixed with a solution of ligand  $\text{L}'$  (0.450 g, 2.29 mmol) in hot water-methanol (1.3 v/v, 30 ml) and the mixture refluxed (5h) on a steam bath. The resulting yellow-green precipitate was filtered hot, washed with hot aq. methanol and dried over fused  $\text{CaCl}_2$  in a dessicator. Yield : 40% m.p. > 300°C.

*Analy. data* : found % (calc. for  $(\text{VO})_2(\text{OH})_2\text{L}'_2(\text{SO}_4)_4\text{H}_2\text{O}$ ) M 14.04 (14.01), C 21.65 (19.78), H 3.22 (3.57), N 22.43 (23.08).  $\Lambda_M$  ( $\Omega^{-1} \text{cm}^2 \text{mol}^{-1}$ , DMSO) : 47.0  $\mu_{\text{eff}}$  (RT) : 1.30 BM/V. EPR (RT) :  $g_{\text{av}} = 1.96$  IR (KBr),  $\text{cm}^{-1}$  : 3000-3500 (br), 1650 (intensity largely reduced), 1100 (s) 610 (s), 450-460 (ms), 970 (s); and UV-Vis (nujol-mull) : 23530 (w)  $\text{cm}^{-1}$ .

*Complex 8* : A solution of  $\text{UO}_2(\text{CH}_3\text{COO})_2\text{H}_2\text{O}$  (0.210 g, ~0.5 mmol) in hot water-methanol (1:3 v/v, 20 ml) was mixed with solution of ligand  $\text{L}'$  (0.200 g, ~1.02 mmol) in same hot solvent mixture (30 ml) and the contents refluxed (2h) on a steam bath. The resulting red compound formed was filtered hot, washed with hot methanol and dried over fused  $\text{CaCl}_2$  in a desiccator. Yield 60%, m.p. > 300°C

*Analy. data* : found % (calc. for  $\text{UO}_2(\text{Pz-3-CONHNH}_2\text{-2COO})_2$ ) M 37.67 (37.66), C 22.45 (22.78), H 1.68 (1.58), N 16.41 (17.72).  $\Lambda_M$  ( $\Omega^{-1} \text{cm}^2 \text{mol}^{-1}$ , DMSO) : 30.8, IR (KBr),  $\text{cm}^{-1}$  : 1330 (s), 1590 (s), 1130 (ms), 450-460 (ms); and UV-Vis (nujol-mull) : 22730 (br)  $\text{cm}^{-1}$ ,  $^1\text{H}$  NMR,  $\delta$ (ppm) : 942-1024 (Pz-ring), 11.97 (NH).

## Results and Discussion

Conductivity values of  $10^{-3}$  M solution in DMSO for complexes other than 1, 2 and 5 are less for 1:1 electrolytes and suggest their nonelectrolytic behaviour despite some solvolysis. Molecular weight measurements indicate complexes 3, 6 and 7 are dimers.

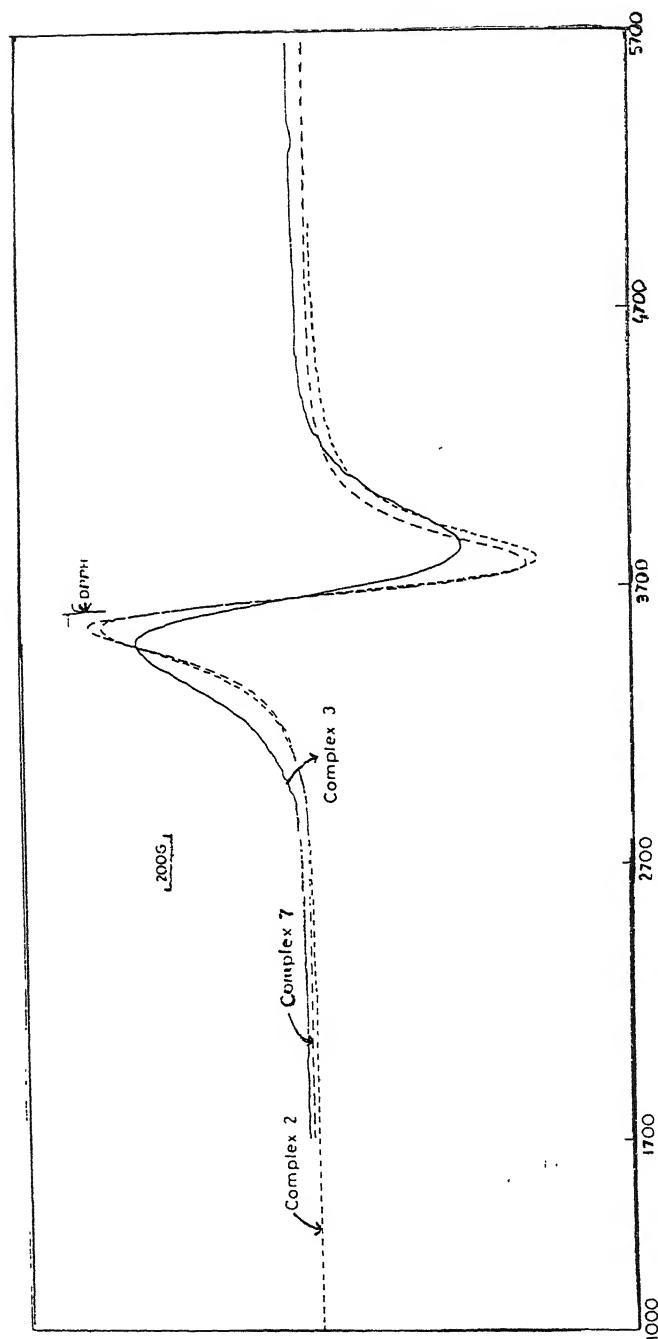


Fig. 4—EPR spectra (RT) frequency 9.45GHz. Field set 3000G for complex 2 and 3700G for complexes 3&7.

*Infrared spectra :* Pyrazine-2-carboxylic acid hydrazide (L) shows bands at 1675 and 1020  $\text{cm}^{-1}$  tentatively assigned<sup>11</sup> to  $\nu\text{C=O}$  and  $\nu\text{N-N}$  respectively; these suffered negative and positive shifts ( $\Delta\nu \sim 20 \text{ cm}^{-1}$ ) in complexes 1 and 2 suggesting coordination through the carboxyl oxygen and the terminal hydrazinic nitrogen. Bands observed for 2 in the region 1240 (w, sh), 1170-1139, 1050-1030  $\text{cm}^{-1}$  indicate<sup>12</sup> sulphate is bidentately chelated. In complex 3, disappearance for  $\nu\text{C=O}$  and appearance of a very strong band to around 1580  $\text{cm}^{-1}$  are characteristic of  $\text{>C=N-N=C<}$  group. A new band at  $\sim 1420 \text{ cm}^{-1}$  (absent in the free ligand) may be assigned<sup>12</sup> to deformation mode of methyl group derived from 'acac' moiety. Non-ligand bands at 530(s) and 840  $\text{cm}^{-1}$  (m, sh) may be attributed<sup>13</sup> to O-V-O sym and asym stretching modes of the oxobridged portion of the dimer. In complex 4,  $\nu\text{C=O}$  band disappeared while a strong band appeared at  $\sim 1600 \text{ cm}^{-1}$ . This along with a new band at 1260  $\text{cm}^{-1}$  suggests<sup>1</sup> coordination through the deprotonated enol oxygen. Positive shift ( $\Delta\nu \sim 20 \text{ cm}^{-1}$ ) of  $\nu\text{N-N}$  band in 4 points to coordination through the terminal hydrazinic nitrogen. Absence of N-H signal from its free ligand position at  $\delta$  10 ppm suggests deprotonation via enolisation, and downfield shift of free ligand signal at  $\delta$  4.64 ppm to  $\delta$  5.08 ppm in the complex supports coordination through  $\text{NH}_2$  group of the ligand (Fig. 3).

In complexes 5 and 7, the strong  $\nu\text{C=O}$  band at 1650  $\text{cm}^{-1}$  of the free dihydrazide ligand ( $\text{L}'$ ) is reduced in intensity (though not shifted); coordination of keto oxygen to the metal is indicated. Band attributable<sup>12</sup> to C–O stretching of coordinated methoxide in 5 at around 1000  $\text{cm}^{-1}$  is not, however, discernible from the complexity of spectra. Sulphate is ionic in 7 apparent from the strong band around 1110  $\text{cm}^{-1}$  and another at 610 (ms)  $\text{cm}^{-1}$ . In complexes 5-8, coordination through heterocyclic ring nitrogen is suggested from the observed shift of pyrazine ring out-of-plane deformation band to higher frequencies (450-460  $\text{cm}^{-1}$ ) relative to the free ligand (400, 415  $\text{cm}^{-1}$ ). Downfield shift of protons of pyrazine ring from the region  $\delta$  8.63 – 9.08 to  $\delta$  9.42 – 10.24 ppm in the  $^1\text{H}$  NMR spectrum of 8 indicates coordination of pyrazine ring nitrogen to the metal : the signal at  $\delta$  11.97 ppm is ascribed to NH (perhaps hydrogen bonded) of a free hydrazide residue. Complexes 6 and 7 which are dimers are possibly hydroxo-bridged, a new band (ms) around 560-580  $\text{cm}^{-1}$  is assignable<sup>14</sup> to  $\nu(\text{V-O})$  of the  $\mu$ -hydroxobridges. A strong characteristic band for complexes 6 and 8 at  $\sim 1330 \text{ cm}^{-1}$  (absent in the free ligand) is perhaps due to  $\nu_{\text{as}}\text{COO}$ ; the corresponding  $\nu_{\text{as}}\text{COO}$  may be assigned to the strong band at  $\sim 1590 \text{ cm}^{-1}$ . The complexes show a free amide I band (strong) at  $\sim 1650 \text{ cm}^{-1}$ ; presence of a free hydrazide group is evident<sup>11</sup> from  $\text{NH}_2$  rocking mode  $\sim 1130 \text{ cm}^{-1}$ .

In all the complexes strong band assignable to  $\nu\text{V=O}$  around 970-980  $\text{cm}^{-1}$ ; and to  $\nu_{\text{as}}\text{O=U=O}$  in the region 880-910  $\text{cm}^{-1}$  have been observed. Non-ligand bands occurring in the regions 500, 550 and 370  $\text{cm}^{-1}$  may be assigned<sup>12</sup>, respectively, to U–O and U–N

vibrations. Broad band observed in the region 3000-3500  $\text{cm}^{-1}$  for complexes 1, 2, 5 and 7 suggest presence of water and/or hydrogen bonding. Isothermal (110°C, air oven) mass loss (9.57, 7.19 and 10.6%) for the corresponding complexes 2, 5 and 7 suggests crystalline water entirely and that (11.5%) for 1 partly (2 mols).

Magnetic moment ( $\mu_{\text{eff}}$ ) values observed at room temperature (33°C) for the oxovanadium (IV) complexes (1, 2 and 5) are normal for magnetically dilute species. Complexes 3, 6 and 7 give lower values similar to those observed<sup>1,15</sup> elsewhere, lowering of magnetic moment in these dimers may indicate<sup>15</sup> antiferromagnetic coupling between the vanadium centers since fortunately, spin-orbit coupling in nearly all V(IV) complexes is efficiently quenched<sup>16</sup>.

EPR spectra of the oxovanadium (IV) complexes in polycrystalline state at room temperature (Fig. 4) are nearly isotropic and the  $g_{\text{av}}$  values are close to 1.96-1.97. No half-filled transition has been located in polycrystalline solids at room temperature even at high gain (however, in absence of epr study at low temperature and/or variable temperature magnetic measurements, which are inaccessible to us, results remain inconclusive).

Electronic spectra of the oxovanadium (IV) complexes in nujol mull exhibit bands in the region 400-440 and 500-600 nm attributable<sup>15</sup>, respectively to  $^2B_2(d_{xy}) \rightarrow ^2A_1(d_{z2})$ ,  $^2B_2(d_{xy}) \rightarrow ^2B_1(d_{x2-y2})$  transitions.

Non-ligand bands observed for complexes 4 and 8 in the region 430-400 nm may be assigned to the  $^1\sum_g^+ \rightarrow ^3\Pi_u$  transition of the  $\text{UO}_2$  moiety<sup>17</sup>.

#### Acknowledgements

The authors thank the authorities of CDRI, Lucknow, for FAB-MS and RSIC, Bose Institute, Kolkata, for EPR measurements. One of the authors (RG) thanks University of Kalyani for the award of Fellowship.

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# Reflection and transmission of $P$ and $SV$ waves at the interface between two monoclinic elastic half-spaces

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Received December 6, 1999; Accepted April 2, 2000

## Abstract

Reflection and transmission of quasi  $-P$  ( $qP$ ) and quasi  $-SV$  ( $qSV$ ) waves at the interface between two monoclinic elastic half-spaces is discussed. Closed-form expressions for the reflection and transmission coefficients are derived. A method of computing these coefficients is indicated. The present analysis corrects some fundamental errors appearing in a recent paper on the reflection and transmission of  $qP$  waves at the interface between two monoclinic media.

(Keywords : reflection/transmission/waves/monoclinic media)

## Introduction

Musgrave<sup>1</sup> discussed the reflection and transmission of elastic waves at a plane boundary between two anisotropic media of hexagonal type. Dayley and Hron<sup>2</sup> investigated the case when the media involved are transversely isotropic. Keith and Crampin<sup>3</sup> derived a formulation for computing the energy division among  $qP$ ,  $qSV$  and  $qSH$  waves generated by plane waves incident on a plane boundary between generally anisotropic media. A comprehensive account was presented for the case of isotropic/orthotropic interface.

A monoclinic medium possesses one plane of elastic symmetry. For wave propagation in the plane of symmetry,  $SH$  motion is decoupled from the  $P$ - $SV$  motion. While the particle motion of  $SH$  waves is purely transverse, it is neither purely longitudinal nor purely transverse in the case of  $P$ - $SV$  waves. In a recent paper, Chattopadhyay and Saha<sup>4</sup> discussed the reflection of  $qP$  waves at the interface between two monoclinic half-spaces. Since the authors assume that  $qP$  waves are purely longitudinal and  $qSV$  waves purely transverse, most of the results of this paper, including the expressions for the reflection and transmission coefficients, are erroneous (see also Singh<sup>5</sup>). The aim of the present study is to derive closed-form algebraic expressions for the reflection and transmission coefficients when plane waves of  $qP$  or  $qSV$  type are incident at the plane boundary between two monoclinic elastic half-spaces. A method of computing the reflection and transmission coefficients is indicated. Numerical results will be presented in a subsequent publication.

### Plane Waves in a Monoclinic Elastic Medium

Consider a homogeneous anisotropic elastic medium of monoclinic type. It has one plane of elastic symmetry and its elastic properties are defined by thirteen elastic moduli. Taking the plane of symmetry as the  $x_2x_3$ -plane, the generalized Hooke's law can be expressed in the form

$$\tau_{11} = c_{11} e_{11} + c_{12} e_{22} + c_{13} e_{33} + 2c_{14} e_{23}, \quad (1a)$$

$$\tau_{22} = c_{12} e_{11} + c_{22} e_{22} + c_{23} e_{33} + 2e_{24} e_{23}, \quad (1b)$$

$$\tau_{33} = c_{13} e_{11} + c_{23} e_{22} + c_{33} e_{33} + 2c_{34} e_{23}, \quad (1c)$$

$$\tau_{23} = c_{14} e_{11} + c_{24} e_{22} + c_{34} e_{33} + 2c_{44} e_{23}, \quad (1d)$$

$$\tau_{13} = 2(c_{55} e_{13} + c_{56} e_{12}), \quad (1e)$$

$$\tau_{12} = 2(c_{56} e_{13} + c_{66} e_{12}), \quad (1f)$$

where  $\tau_{ij}$  is the stress tensor and  $e_{ij}$  the strain tensor. Further,

$$2e_{ij} = \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}, \quad (2)$$

$u_i$  being the displacement vector.

For plane waves propagating in the  $x_2x_3$ -plane

$$u_i = u_i(x_2, x_3, t), \quad \partial/\partial x_1 \equiv 0. \quad (3)$$

The equations of motion without body forces are

$$\frac{\partial}{\partial x_j} \tau_{ij} = \rho \frac{\partial^2 u_i}{\partial t^2} \quad (i = 1, 2, 3), \quad (4)$$

using the summation convention. From eqn. (1) to (4), we obtain the equations of motion in terms of the displacements in the form

$$c_{66} \frac{\partial^2 u_1}{\partial x_2^2} + 2c_{56} \frac{\partial^2 u_1}{\partial x_2 \partial x_3} + c_{55} \frac{\partial^2 u_1}{\partial x_3^2} = \rho \frac{\partial^2 u_1}{\partial t^2}, \quad (5)$$

$$\begin{aligned} c_{22} \frac{\partial^2 u_2}{\partial x_2^2} + c_{44} \frac{\partial^2 u_2}{\partial x_3^2} + c_{24} \frac{\partial^2 u_3}{\partial x_2^2} + c_{34} \frac{\partial^2 u_3}{\partial x_3^2} + 2c_{24} \frac{\partial^2 u_2}{\partial x_2 \partial x_3} \\ + (c_{23} + c_{44}) \frac{\partial^2 u_3}{\partial x_2 \partial x_3} = \rho \frac{\partial^2 u_2}{\partial t^2}, \end{aligned} \quad (6)$$

$$\begin{aligned} c_{24} \frac{\partial^2 u_2}{\partial x_2^2} + c_{34} \frac{\partial^2 u_2}{\partial x_3^2} + c_{44} \frac{\partial^2 u_3}{\partial x_2^2} + c_{33} \frac{\partial^2 u_3}{\partial x_3^2} + 2c_{34} \frac{\partial^2 u_3}{\partial x_2 \partial x_3} \\ + (c_{23} + c_{44}) \frac{\partial^2 u_2}{\partial x_2 \partial x_3} = \rho \frac{\partial^2 u_3}{\partial t^2}. \end{aligned} \quad (7)$$

From eqn. (5) to (7), it is obvious that the  $u_1$  motion representing *SH* waves is decoupled from the  $(u_2, u_3)$  motion representing *qP* and *qSV* waves.

Let  $\mathbf{p}(0, p_2, p_3)$  denote the unit propagation vector,  $c$  the phase velocity and  $k$  the wave number of plane waves propagating in the  $x_2 x_3$ -plane. We seek plane wave solutions of the equations of motion (6) and (7) of the form

$$\begin{pmatrix} u_2 \\ u_3 \end{pmatrix} = A \begin{pmatrix} d_2 \\ d_3 \end{pmatrix} \exp [ik(ct - x_2 p_2 - x_3 p_3)], \quad (8)$$

where  $\mathbf{d}(0, d_2, d_3)$  is the unit displacement vector, also known as the polarization vector. Inserting the expressions for  $u_2$  and  $u_3$  in the equations of motion (6) and (7), we obtain

$$(U - \rho c^2) d_2 + V d_3 = 0, \quad (9)$$

$$V d_2 + (Z - \rho c^2) d_3 = 0, \quad (10)$$

where

$$\begin{aligned} U(p_2, p_3) &= c_{22} p_2^2 + c_{44} p_3^2 + 2c_{24} p_2 p_3, \\ V(p_2, p_3) &= c_{24} p_2^2 + c_{34} p_3^2 + (c_{23} + c_{44}) p_2 p_3, \\ Z(p_2, p_3) &= c_{44} p_2^2 + c_{33} p_3^2 + 2c_{34} p_2 p_3. \end{aligned} \quad (11)$$

Eqn. (9) and (10) yield

$$d_2 / d_3 = V / (\rho c^2 - U) = (\rho c^2 - Z) / V. \quad (12)$$

Therefore,  $\rho c^2$  satisfies the quadratic equation

$$\rho^2 c^4 - (U + Z) \rho c^2 + (UZ - V^2) = 0, \quad (13)$$

with solutions

$$2\rho c^2 (p_2, p_3) = (U + Z) \pm [(U - Z)^2 + 4V^2]^{1/2}. \quad (14)$$

The upper sign in eqn. (14) is for  $qP$  waves and the lower sign is for  $qSV$  waves.

It has been shown by Singh<sup>5</sup> that eqn. (8) will represent a pure longitudinal or transverse wave if

$$\begin{aligned} c_{24} p_2^4 + (c_{23} - c_{22} + 2c_{44}) p_2^3 p_3 - 3(c_{24} - c_{34}) p_2^2 p_3^2 \\ - (c_{23} - c_{33} + 2c_{44}) p_2 p_3^3 - c_{34} p_3^4 = 0. \end{aligned} \quad (15)$$

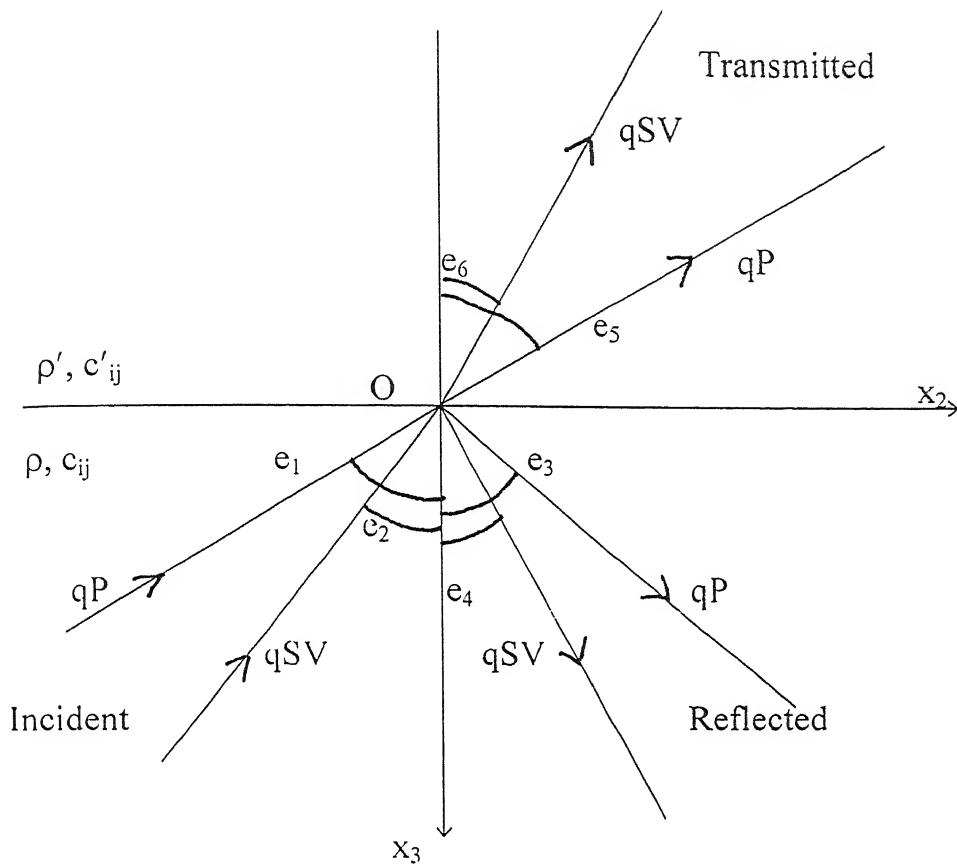


Fig. 1 – Reflection and transmission of  $qP$  and  $qSV$  waves at the plane interface ( $x_3 = 0$ ) between two monoclinic half-spaces.

Eqn. (15) gives the directions of propagation for which  $P$  waves are purely longitudinal and  $SV$  waves purely transverse.

#### Reflection and Transmission of $qP$ and $qSV$ Waves

Consider a homogeneous, monoclinic, elastic half-space occupying the region  $x_3 > 0$  in welded contact with another homogeneous, monoclinic, elastic half-space  $x_3 < 0$  (Fig. 1). The identical plane of elastic symmetry of the two media is taken as the  $x_2x_3$  – plane. Plane  $qP$  or  $qSV$  waves are incident at the interface  $x_3 = 0$  from the half-space  $x_3 > 0$ . We consider plane strain problem for which

$$u_1 = 0, \quad u_2 = u_2(x_2, x_3, t), \quad u_3 = u_3(x_2, x_3, t). \quad (16)$$

Incident  $qP$  or  $qSV$  waves will generate reflected  $qP$  and  $qSV$  waves in the half-space  $x_3 > 0$  and transmitted  $qP$  and  $qSV$  waves in the half-space  $x_3 < 0$ . The total displacement field is given by

$$u_2 = \sum_{j=1}^4 A_j e^{iP_j}, \quad u_3 = \sum_{j=1}^4 B_j e^{iP_j} \quad (17)$$

for  $x_3 > 0$ , and

$$u_2' = \sum_{j=5}^6 A_j e^{iP_j}, \quad u_3' = \sum_{j=5}^6 B_j e^{iP_j} \quad (18)$$

for  $x_3 < 0$ , where

$$P_j = \omega [t - (x_2 \sin e_j - x_3 \cos e_j) / c_j], \quad (j = 1, 2, 5, 6; \text{ no summation}) \quad (19)$$

$$P_j = \omega [t - (x_2 \sin e_j + x_3 \cos e_j) / c_j], \quad (j = 3, 4) \quad (20)$$

$\omega$  being the angular frequency. We distinguish quantities corresponding to various waves by using the subscript (1) for incident  $qP$  waves, (2) for incident  $qSV$  waves, (3) for reflected  $qP$  waves, (4) for reflected  $qSV$  waves, (5) for transmitted  $qP$  waves and (6) for transmitted  $qSV$  waves. Thus, for example, for the incident  $qP$  waves,  $c_1$  denotes the phase velocity,  $e_1$  the angle of incidence,  $P_1(x_2, x_3, t)$  the phase factor,  $A_1$  the amplitude factor of the  $u_2$  component of the displacement and  $B_1$  that of the  $u_3$  component. The displacement components in the half-space  $x_3 < 0$  are denoted by  $u_2'$  and  $u_3'$ .

Since each of the incident  $qP$ , incident  $qSV$ , reflected  $qP$ , reflected  $qSV$ , transmitted  $qP$  and transmitted  $qSV$  waves must satisfy the equations of motion, we have, as in eqn. (12) and (14),

$$A_i = F_i B_i \quad (i = 1, 2, \dots, 6), \quad (21)$$

where

$$F_i = V_i / (\rho c_i^2 - U_i) = (\rho c_i^2 - Z_i) / V_i, \quad (i = 1, 2, 3, 4), \quad (22a)$$

$$F_i = V_i / (\rho' c_i^2 - U_i) = (\rho' c_i^2 - Z_i) / V_i, \quad (i = 5, 6), \quad (22b)$$

$$2\rho c_i^2 = (U_i + Z_i) + [(U_i - Z_i)^2 + 4V_i^2]^{1/2}, \quad (i = 1, 3), \quad (23a)$$

$$2\rho c_i^2 = (U_i + Z_i) - [(U_i - Z_i)^2 + 4V_i^2]^{1/2}, \quad (i = 2, 4) \quad (23b)$$

$$2\rho' c_5^2 = U_5 + Z_5 + [(U_5 - Z_5)^2 + 4V_5^2]^{1/2}, \quad (24a)$$

$$2\rho' c_6^2 = U_6 + Z_6 - [(U_6 - Z_6)^2 + 4V_6^2]^{1/2}. \quad (24b)$$

The expressions for  $U_i$ ,  $V_i$  and  $Z_i$  are obtained from the expressions for  $U$ ,  $V$  and  $Z$  given in eqn. (11) on substituting suitable values for  $(p_2, p_3)$ . For incident *qP* waves,  $p_2 = \sin e_1$ ,  $p_3 = -\cos e_1$ ; for incident *qSV* waves,  $p_2 = \sin e_2$ ,  $p_3 = -\cos e_2$ ; for reflected *qP* waves,  $p_2 = \sin e_3$ ,  $p_3 = \cos e_3$ ; for reflected *qSV* waves,  $p_2 = \sin e_4$ ,  $p_3 = \cos e_4$ ; for transmitted *qP* waves,  $p_2 = \sin e_5$ ,  $p_3 = -\cos e_5$ ; and, for transmitted *qSV* waves,  $p_2 = \sin e_6$ ,  $p_3 = -\cos e_6$  (see Fig. 1). We thus obtain

$$U_1 = c_{22} \sin^2 e_1 + c_{44} \cos^2 e_1 - 2c_{24} \sin e_1 \cos e_1,$$

$$V_1 = c_{24} \sin^2 e_1 + c_{34} \cos^2 e_1 - (c_{23} + c_{44}) \sin e_1 \cos e_1,$$

$$Z_1 = c_{44} \sin^2 e_1 + c_{33} \cos^2 e_1 - 2c_{34} \sin e_1 \cos e_1; \quad (25)$$

$$U_3 = c_{22} \sin^2 e_3 + c_{44} \cos^2 e_3 + 2c_{24} \sin e_3 \cos e_3,$$

$$V_3 = c_{24} \sin^2 e_3 + c_{34} \cos^2 e_3 + (c_{23} + c_{44}) \sin e_3 \cos e_3,$$

$$Z_3 = c_{44} \sin^2 e_3 + c_{33} \cos^2 e_3 + 2c_{34} \sin e_3 \cos e_3; \quad (26)$$

$$U_5 = c'_{22} \sin^2 e_5 + c'_{44} \cos^2 e_5 - 2c'_{24} \sin e_5 \cos e_5,$$

$$V_5 = c'_{24} \sin^2 e_5 + c'_{34} \cos^2 e_5 - (c'_{23} + c'_{44}) \sin e_5 \cos e_5,$$

$$Z_5 = c'_{44} \sin^2 e_5 + c'_{33} \cos^2 e_5 - 2c'_{34} \sin e_5 \cos e_5. \quad (27)$$

$(U_2, V_2, Z_2)$  are obtained from  $(U_1, V_1, Z_1)$  on replacing  $e_1$  by  $e_2$ ,  $(U_4, V_4, Z_4)$  are obtained from  $(U_3, V_3, Z_3)$  on replacing  $e_3$  by  $e_4$  and  $(U_6, V_6, Z_6)$  are obtained from  $(U_5, V_5, Z_5)$  on replacing  $e_5$  by  $e_6$ .

The total displacement field given by eqn. (17) and (18) must satisfy the boundary conditions,

$$u_2 = u'_2, u_3 = u'_3, \tau_{23} = \tau'_{23}, \tau_{33} = \tau'_{33} \text{ at } x_3 = 0. \quad (28)$$

Since the boundary conditions (28) are to be satisfied for all values of  $x_2$ , we must have

$$P_1(x_2, 0) = P_2(x_2, 0) = P_3(x_2, 0) = P_4(x_2, 0) = P_5(x_2, 0) = P_6(x_2, 0). \quad (29)$$

Eqn. (19), (20) and (29) imply

$$\frac{\sin e_1}{c_1(e_1)} = \frac{\sin e_2}{c_2(e_2)} = \frac{\sin e_3}{c_3(e_3)} = \frac{\sin e_4}{c_4(e_4)} = \frac{\sin e_5}{c_5(e_5)} = \frac{\sin e_6}{c_6(e_5)} = 1/c_a, \quad (30)$$

where  $c_a$  is the apparent phase velocity. This is the form of Snell's law for monoclinic media.

From eqn. (3a), (25) and (26), we note that even if  $e_1 = e_3$ ,  $c_1 \neq c_3$ . Therefore, from eqn. (30), the angle of reflection of  $qP$  waves is not equal to the angle of incidence of  $qP$  waves. Similarly, the angle of reflection of  $qSV$  waves is not equal to the angle of

idence of  $qSV$  waves. Chattopadhyay and Saha<sup>4</sup> assume that the angle of reflection of  $(qSV)$  waves is equal to the angle of incidence of  $qP$  ( $qSV$ ) waves. Therefore, the flection and the transmission coefficients obtained by Chattopadhyay and Saha<sup>4</sup> are not correct.

Using the relations (21), (29) and (30) the bounday conditions (28) yield

$$B_1 + B_2 + B_3 + B_4 - B_5 - B_6 = 0, \quad (31a)$$

$$F_1 B_1 + F_2 B_2 + F_3 B_3 + F_4 B_4 - F_5 B_5 - F_6 B_6 = 0, \quad (31b)$$

$$a_1 B_1 + a_2 B_2 + a_3 B_3 + a_4 B_4 - a_5 B_5 - a_6 B_6 = 0, \quad (31c)$$

$$b_1 B_1 + b_2 B_2 + b_3 B_3 + b_4 B_4 - b_5 B_5 - b_6 B_6 = 0, \quad (31d)$$

here

$$a_1 = c_{24} F_1 + c_{44} - (c_{44} F_1 + c_{34}) \cot e_1,$$

$$a_2 = c_{24} F_2 + c_{44} - (c_{44} F_2 + c_{34}) \cot e_2,$$

$$a_3 = c_{24} F_3 + c_{44} + (c_{44} F_3 + c_{34}) \cot e_3,$$

$$a_4 = c_{24} F_4 + c_{44} + (c_{44} F_4 + c_{34}) \cot e_4,$$

$$a_5 = c'_{24} F_5 + c'_{44} - (c'_{44} F_5 + c'_{34}) \cot e_5,$$

$$a_6 = c'_{24} F_6 + c'_{44} - (c'_{44} F_6 + c'_{34}) \cot e_6,$$

$$b_1 = c_{23} F_1 + c_{34} - (c_{34} F_1 + c_{33}) \cot e_1,$$

$$b_2 = c_{23} F_2 + c_{34} - (c_{34} F_2 + c_{33}) \cot e_2,$$

$$b_3 = c_{23} F_3 + c_{34} + (c_{34} F_3 + c_{33}) \cot e_3,$$

$$b_4 = c_{23}F_4 + c_{34} + (c_{34}F_4 + c_{33}) \cot e_4,$$

$$b_5 = c'_{23}F_5 + c'_{34} - (c'_{34}F_5 + c'_{33}) \cot e_5,$$

$$b_6 = c'_{23}F_6 + c'_{34} - (c'_{34}F_6 + c'_{33}) \cot e_6.$$

*Incident qP waves :*

In the case of incident *qP* waves,  $A_2 = B_2 = 0$  and  $A_1, B_1$  are supposed to be known. Eqn. (31a, b, c, d) then constitute a set of four simultaneous equations in four unknowns, namely,  $B_3, B_4, B_5$  and  $B_6$ . These equations can be solved by Cramer's rule. We find

$$B_i / B_1 = \Delta_i^P / \Delta \quad (i = 3, 4, 5, 6), \quad (32)$$

where  $\Delta$  and  $\Delta_i^P$  are defined in Appendix A. Using eqn. (21), we find

$$\frac{A_i}{A_1} = \frac{F_i}{F_1} \left( \frac{B_i}{B_1} \right) = \frac{F_i}{F_1} \left( \frac{\Delta_i^P}{\Delta} \right) \quad (i = 3, 4, 5, 6; \text{ no summation over } i). \quad (33)$$

*Incident qSV waves :*

For incident *qSV* waves,  $A_1 = B_1 = 0$  and  $A_2, B_2$  are supposed to be known. The amplitude ratios are found to be

$$B_i / B_2 = \Delta_i^S / \Delta, \quad (34)$$

$$\frac{A_i}{A_2} = \frac{F_i}{F_2} \left( \frac{B_i}{B_2} \right) = \frac{F_i}{F_2} \left( \frac{\Delta_i^S}{\Delta} \right) \quad (i = 3, 4, 5, 6), \quad (35)$$

where  $\Delta_i^S$  are defined in Appendix A.

*Isotropic half-spaces :*

For an isotropic medium,

$$c_{11} = c_{22} = c_{33} = \lambda + 2\mu,$$

$$c_{12} = c_{13} = c_{23} = \lambda, \quad c_{44} = c_{55} = c_{66} = \mu, \\ c_{14} = c_{24} = c_{34} = c_{56} = 0, \quad (36)$$

where  $\lambda, \mu$  are the Lamé parameters. Using these values for  $c_{ij}$  and similar values for  $c'_{ij}$  we obtain

$$c_1 = c_3 = [\lambda + 2\mu]^{1/2} = \alpha, \quad c_2 = c_4 = (\mu / \rho)^{1/2} = \beta,$$

$$c_5 = [(\lambda' + 2\mu') / \rho']^{1/2} = \alpha', \quad c_6 = (\mu' / \rho')^{1/2} = \beta',$$

$$e_1 = e_3 = e, \quad e_2 = e_4 = f, \quad e_5 = e', \quad e_6 = f',$$

$$\frac{\sin e}{\alpha} = \frac{\sin f}{\beta} = \frac{\sin e'}{\alpha'} = \frac{\sin f'}{\beta'},$$

$$F_1 = -F_3 = -\tan e, \quad F_2 = -F_4 = \cot f, \quad F_5 = -\tan e', \quad F_6 = \cot f',$$

$$a_1 = a_3 = 2\mu, \quad a_2 = a_4 = -\mu \cos 2f / \sin^2 f, \quad a_5 = 2\mu', \quad a_6 = -\mu' \cos 2f' / \sin^2 f',$$

$$b_1 = -b_3 = -2\mu (\alpha/\beta)^2 \cos 2f / \sin 2e,$$

$$b_2 = -b_4 = -2\mu \cot f, \quad b_5 = -2\mu' (\alpha'/\beta')^2 \cos 2f' / \sin 2e', \quad b_6 = -2\mu' \cot f' \quad (37)$$

Putting these values in eqn. (31a, b, c, d), we get results equivalent to the corresponding results given by Ben-Menahem and Singh<sup>6</sup> (eqn. (3.54) and (3.56)) for isotropic media.

### Discussion and Conclusions

The reflection and transmission coefficients given by Chattopadhyay and Saha<sup>4</sup> for *qP* waves incident at the plane boundary between two monoclinic elastic half-spaces are incorrect because of two erroneous assumptions made by these authors, namely, *qP* waves are longitudinal (*qSV* waves are transverse) and the angle of reflection of *qP* (*qSV*) waves

is equal to the angle of incidence of  $qP$  ( $qSV$ ) waves. In the present study, we have obtained the correct reflection and transmission coefficients by solving the problem *ab initio*.

Eqn. (32) and (33) give the amplitude ratios when plane  $qP$  waves are incident at the plane boundary between two monoclinic elastic half-spaces. In these equations,  $A_i/A_1$  are the amplitude ratios for the horizontal component of the displacement and  $B_i/B_1$  are the amplitude ratios for the vertical component of the displacement. Similarly, eqn. (34) and (35) give the amplitude ratios for incident  $qSV$  waves. From eqn. (17) and (21), we note that, for example, the total displacement of the incident  $qP$  waves is

$$(A_1^2 + B_1^2)^{1/2} e^{iP_1} = (1 + F_1^2)^{1/2} B_1 e^{iP_1}.$$

Therefore, the reflection coefficients can be expressed in the form

$$R_{PP} = \left( \frac{1 + F_3^2}{1 + F_1^2} \right)^{1/2} \cdot \frac{B_3}{B_1}, \quad R_{PS} = \left( \frac{1 + F_4^2}{1 + F_1^2} \right)^{1/2} \cdot \frac{B_4}{B_1} \quad (38)$$

for incident  $qP$  waves, and

$$R_{SP} = \left( \frac{1 + F_3^2}{1 + F_2^2} \right)^{1/2} \cdot \frac{B_3}{B_2}, \quad R_{SS} = \left( \frac{1 + F_4^2}{1 + F_2^2} \right)^{1/2} \cdot \frac{B_4}{B_2} \quad (39)$$

for incident  $qSV$  waves. Similar expressions can be written for the transmission coefficients. The reflection and transmission coefficients are in terms of the six angles  $e_i$  and the six velocities  $c_i(e_i)$ ,  $i = 1, 2, \dots, 6$ . For an incident  $qP$  wave,  $e_1$  and, therefore,  $c_1(e_1)$  is supposed to be known. One has to compute  $e_i$  ( $i = 3, 4, 5, 6$ ) for given  $e_1$ . The velocities  $c_i(e_i)$  can then be computed from explicit algebraic formulae. We give below the procedure for computing  $e_i$  for given  $e_1$  in the case of incident  $qP$  waves and for given  $e_2$  in the case of incident  $qSV$  waves.

The Snell's law for a monoclinic medium is given by eqn. (30) in which the apparent velocity  $c_a$  can be written as  $c_a = c/p_2$ , where  $p$  ( $0, p_2, p_3$ ) is the propagation vector. We define dimensionless apparent velocity  $c$  through the relation

$$\bar{c} = c_d/\beta = c/(p_2\beta), \quad (40)$$

where  $\beta = (c_{44}/\rho)^{1/2}$ . Eqn. (13) then becomes

$$\bar{c}^4 - (\bar{U} + \bar{Z}) \bar{c}^2 + (\bar{U}\bar{Z} - \bar{V}^2) = 0, \quad (41)$$

where

$$\bar{U} = U/(c_{44}p_2^2) = p^2 + 2\bar{c}_{24}p + \bar{c}_{22},$$

$$\bar{V} = V/(c_{44}p_2^2) = \bar{c}_{34}p^2 + (1 + \bar{c}_{23})p + \bar{c}_{24},$$

$$\bar{Z} = Z/(c_{44}p_2^2) = \bar{c}_{33}p^2 + 2\bar{c}_{33}p + 1,$$

$$p = p_3/p_2, \quad \bar{c}_{ij} = c_{ij}/c_{44}. \quad (42)$$

For incident  $qP$  waves,  $p = -\cot e_1$ ; for incident  $qSV$  waves,  $p = -\cot e_2$ ; for reflected  $qP$  waves,  $p = \cot e_3$ ; for reflected  $qSV$  waves,  $p = \cot e_4$ ; for transmitted  $qP$  waves,  $p = -\cot e_5$ ; for transmitted  $qSV$  waves,  $p = -\cot e_6$ . For a given  $p$ , eqn. (41) can be solved for  $\bar{c}^2$ , the two roots corresponding to  $qP$  and  $qSV$  waves. However, for a given  $\bar{c}$ , eqn. (41) is a bi-quadratic in  $p$ , corresponding to incident  $qP$ , incident  $qSV$ , reflected  $qP$  and reflected  $qSV$ . The positive roots corresponding to the reflected waves and the negative roots corresponding to the incident waves. On inserting the expressions for  $\bar{U}$ ,  $\bar{Z}$  and  $\bar{V}$  from eqn. (42) into eqn. (41), the bi-quadratic in  $p$  becomes

$$g_0 p^4 + g_1 p^3 + g_2 p^2 + g_3 p + g_4 = 0, \quad (43)$$

where  $g_0 = \bar{c}_{33} - \bar{c}_{34}^2$ ,

$$g_1 = 2(\bar{c}_{24} \bar{c}_{33} - \bar{c}_{23} \bar{c}_{34}),$$

$$\begin{aligned}
 g_2 &= 1 + \bar{c}_{22} \bar{c}_{33} + 2 \bar{c}_{24} \bar{c}_{34} - (1 + \bar{c}_{23})^2 - (1 + \bar{c}_{33}) \bar{c}^2, \\
 g_3 &= 2[\bar{c}_{22} \bar{c}_{34} - \bar{c}_{23} \bar{c}_{24} - (\bar{c}_{24} + \bar{c}_{34}) \bar{c}^2], \\
 g_4 &= \bar{c}^4 - (1 + \bar{c}_{22}) \bar{c}^2 + \bar{c}_{22} - \bar{c}_{24}^2. \tag{44}
 \end{aligned}$$

If we define  $q = 1/p = p_2/p_3$ , the bi-quadratic transforms to

$$g_4q^4 + g_3q^3 + g_2q^2 + g_1q + g_0 = 0. \tag{45}$$

For angles of incidence, for which both reflected  $qP$  and reflected  $qSV$  waves exist, eqn. (45) will possess two positive roots, the smaller positive root (say  $q_4$ ) corresponding to reflected  $SV$  and the larger positive root ( $q_3$ ) corresponding to reflected  $qP$ . Further,

$$e_3 = \tan^{-1}(q_3), \quad e_4 = \tan^{-1}(q_4). \tag{46}$$

A similar procedure can be set up for finding  $e_5$  and  $e_6$ .

#### Acknowledgements

The authors are grateful to the Council of Scientific and Industrial Research, New Delhi for financial support through the Emeritus Scientist Scheme awarded to SJS and to the University Grants Commission, New Delhi for Junior Research Fellowship awarded to SK.

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## Appendix A

$$\Delta = \begin{vmatrix} 1 & 1 & -1 & -1 \\ F_3 & F_4 & -F_5 & -F_6 \\ a_3 & a_4 & -a_5 & -a_6 \\ b_3 & b_4 & -b_5 & -b_6 \end{vmatrix}$$

$$\Delta_3^P = \begin{vmatrix} -1 & 1 & -1 & -1 \\ -F_1 & F_4 & -F_5 & -F_6 \\ -a_1 & a_4 & -a_5 & -a_6 \\ -b_1 & b_4 & -b_5 & -b_6 \end{vmatrix}$$

$$\Delta_4^P = \begin{vmatrix} 1 & -1 & -1 & -1 \\ F_3 & -F_1 & -F_5 & -F_6 \\ a_3 & -a_1 & -a_5 & -a_6 \\ b_3 & -b_1 & -b_5 & -b_6 \end{vmatrix}$$

$$\Delta_5^P = \begin{vmatrix} 1 & 1 & -1 & -1 \\ F_3 & F_4 & -F_1 & -F_6 \\ a_3 & a_4 & -a_1 & -a_6 \\ b_3 & b_4 & -b_1 & -b_6 \end{vmatrix}$$

$$\Delta_6^P = \begin{vmatrix} 1 & 1 & -1 & -1 \\ F_3 & F_4 & -F_5 & -F_1 \\ a_3 & a_4 & -a_5 & -a_1 \\ b_3 & b_4 & -b_5 & -b_1 \end{vmatrix}$$

$\Delta_3^S$  is obtained from  $\Delta_3^P$  on replacing the elements  $\{-1, -F_1, -a_1, -b_1\}$  in the first column by the elements  $\{-1, -F_2, -a_2, -b_2\}$ .  $\Delta_i^S$  ( $i = 4, 5, 6$ ) are similarly defined.



# Certain generalized subclass of analytic and multivalent functions in terms of fractional calculus

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Received April 1, 2000; Accepted August 29, 2000

## Abstract

Certain generalized subclass of analytic and multivalent functions, with negative coefficients, defined in terms of certain fractional derivative, has been introduced. Present paper investigates results pertinent to coefficient estimate, distortion and inclusion theorems for the said subclass. The class, preserving integral operator of fractional order of certain kind for the generalized subclass, has been investigated too. At the conclusion of this paper, radii of  $p$ -valently starlike and convexity for the said class have been determined.

(Keywords and Phrases : univalent and  $p$ -valent functions/fractional differential and integral operators/convex and starlike convex functions)

## Introduction

Let  $A(p)$  denotes the class of functions of the form

$$f(z) = z^p + \sum_{k=n+p}^{\infty} a_k z^k; \quad n = 1, 2, \dots \quad (1)$$

where  $p$  is a positive integer and  $f(z)$  is analytic and  $p$ -valent in the unit disc  $\mathcal{D} = \{z : |z| < 1\}$ .

Also, let  $T(p)$  be a sub-class of  $A(p)$ , consisting of analytic and  $p$ -valent functions, expressed in the form

$$f(z) = z^p - \sum_{k=n+p}^{\infty} a_k z^k; \quad a_k \geq 0 \quad (2)$$

The function  $f(z) \in T(p)$  is said to be in the subclass  $S^*(p, \delta)$ , iff

$$\operatorname{Re} \left\{ \frac{zf'(z)}{f(z)} \right\} > \delta, \quad z \in \mathcal{D}, \quad (3)$$

for some  $\delta$  ( $0 \leq \delta < p$ ). Function  $f(z) \in S^*(p, \delta)$

$$\operatorname{Re} \left\{ 1 + \frac{zf''(z)}{f'(z)} \right\} > \delta, \quad z \in \mathcal{D} \quad (4)$$

for some  $\delta$  ( $0 \leq \delta < p$ ). Function  $f(z) \in C(p, \delta)$  is called  $p$ -valent convex function of order  $\delta$ . The above said classes  $S^*(p, \delta)$  and  $C(p, \delta)$  were introduced by Owa<sup>1</sup> and studied by Srivastava and Owa<sup>2</sup>.

In what follows, the definition and terminologies of fractional calculus, employed in the present investigations, are due to Owa<sup>3,4</sup>.

*Definition 1.1* : The fractional integral operator of order  $\lambda$ , for a function  $f(z)$ , is defined by

$$D_z^{-\lambda} f(z) = \frac{1}{\Gamma(\lambda)} \int_0^z \frac{f(t)}{(z-t)^{1-\lambda}} dt, \quad \lambda > 0 \quad (5)$$

where  $f(z)$  is the analytic function in a simply-connected region of the  $z$ -plane, containing the origin, and the multiplicity of  $(z-t)^{\lambda-1}$  is removed by requiring  $\log(z-t)$  to be real, when  $(z-t) > 0$ .

*Definition 1.2* : Fractional derivative of order  $\lambda$ , for a function  $f(z)$ , is defined as

$$D_z^\lambda f(z) = \frac{1}{\Gamma(1-\lambda)} \frac{d}{dz} \int_0^z \frac{f(t)}{(z-t)^\lambda} dt, \quad 0 \leq \lambda < 1 \quad (6)$$

while the definition of  $f(z)$  and its validity is given in (5) and multiplicity of  $(z-t)^{\lambda-1}$  is removed as Definition 1.1.

*Definition 1.3* : Under the hypothesis of definition 1.2, the fractional derivative of order  $(n+\lambda)$ , for  $f(z)$ , is expressed as

$$D_z^{n+\lambda} f(z) = \frac{d^n}{dz^n} D_z^\lambda f(z), \quad 0 \leq \lambda < 1 \quad (7)$$

where  $n \in N_0 = N \cup \{0\}$ , where  $N$  is the set of natural numbers.

Let us define the following fractional differential operator  $U_z^{(\lambda, p)}$ ,  $0 \leq \lambda < 1$ .

*Definition 1.4* : For a real number  $\lambda$  ( $0 \leq \lambda < 1$ ), we define

$$U_z^{(\lambda, p)} : T(p) \rightarrow T(p)$$

by

$$U_z^{(\lambda, p)} f(z) = z^p - \sum_{k=n+p}^{\infty} a_k G_p(k, \lambda) z^k, \quad (8)$$

where

$$G_p(k, \lambda) = \frac{\Gamma(k+1) \Gamma(p+1-\lambda)}{\Gamma(p+1) \Gamma(k+1-\lambda)}. \quad (9)$$

For  $z \neq 0$ , we define

$$U_z^{(\lambda, p)} f(z) = \frac{\Gamma(p+1-\lambda)}{\Gamma(p+1)} z^\lambda D_z^\lambda f(z), \quad (10)$$

where  $D_z^\lambda f(z)$  is the fractional derivative of  $f$ , of order  $\lambda$  ( $0 \leq \lambda < 1$ ), while

$$U_z^{(0, p)} f(z) = f(z); \quad U_z^{(1, p)} f(z) = \frac{zf'(z)}{p}. \quad (11)$$

Now, with the help of operator  $U_z^{(\lambda, p)} f(z)$ , we may introduce following class of functions in  $T(p)$ .

*Definition 1.5* : A function  $f(z) \in T(p)$  is said to be in the class  $S_p^*(A, B, \alpha, \beta, \lambda)$ , iff

$$\left| \left\{ \frac{z \left( U_z^{(\lambda, p)} f(z) \right)'}{U_z^{(\lambda, p)} f(z)} - p \right\} / \left\{ (p - \alpha)(A - B) + pB - Bz \frac{(U_z^{(\lambda, p)} f(z))'}{U_z^{(\lambda, p)} f(z)} \right\} \right| < \beta \quad (12)$$

where  $z \in \mathcal{D}$  and its validity is given by  $-1 \leq B < A < 1$ ,  $0 < A \leq 1$ ,  $0 < \beta \leq 1$ ,  $0 \leq \alpha < p$  and  $0 \leq \lambda < 1$ .

The sub-class  $S_p^*(A, B, \alpha, \beta, \lambda)$ , defined by (12) is much general in character and as such number of subclasses become apparent cases of it, which were studied by various authors, we mention some of them

- (i)  $S_p^*(1, -1, \alpha, 1, 0) = S^*(p, \alpha)$  ; (cf. 1)
- (ii)  $S_p^*(1, -1, \alpha, 1, 1) = C(p, \alpha)$  ; (cf. 1)
- (iii)  $S_1^*(1, -1, \alpha, 1, \lambda) = S_\lambda(\alpha)$  ; (cf. 5)
- (iv)  $S_1^*(1, -1, \alpha, \beta, 0) = S(\alpha, \beta)$  ; (cf. 6)
- (v)  $S_1^*(1, -1, \alpha, \beta, 1) = C(\alpha, \beta)$  ; (cf. 6)
- (vi)  $S_p^*(1, 1 - 2\zeta, \alpha, \beta, 0) = S_p^*(\alpha, \beta, \zeta)$  ; (cf. 3) ;  $1/2 < \zeta \leq 1$ .
- (vii)  $S_p^*(A, B, \alpha, 1, 0) = T_p^*(A, B, \alpha)$  ; (cf. 8)
- (viii)  $S_p^*(A, B, \alpha, 1, 1) = C_p(A, B, \alpha)$  ; (cf. 8).

Several other such subclass may be formulated by specializing parameters.

### Coefficients Estimates

*Theorem 1* : A function  $f(z)$ , defined by (2), is in the class  $S_p^*(A, B, \alpha, \beta, \lambda)$ , iff

$$\sum_{k=n+p}^{\infty} \left\{ (1 - \beta B) (k - p) + \beta (A - B) (p - \alpha) \right\} G_p(k, \lambda) a_k \leq (A - B) \beta (p - \alpha), \quad (13)$$

where  $G_p(k, \lambda)$  is given in (9).

The inequality (13) is sharp.

*Proof:* Assume that (13) holds true, and let  $|z| = 1$ , then we have

$$\begin{aligned} & \left| z (U_z^{(\lambda, p)} f(z))' - p U_z^{(\lambda, p)} f(z) \right| - \beta \left| (p - \alpha) (A - B) + p B U_z^{(\lambda, p)} f(z) - B z \left( U_z^{(\lambda, p)} f(z) \right)' \right| \\ &= \left| - \sum_{k=n+p}^{\infty} (k - p) G_p(k, \lambda) a_k z^k \right| \\ & - \beta \left| (A - B) (p - \alpha) z^p + \sum_{k=n+p}^{\infty} \left\{ B (k - p) + \beta (A - B) (p - \alpha) \right\} G_p(k, \lambda) a_k z^k \right| \\ & \leq \sum_{k=n+p}^{\infty} \left| (1 - B\beta) k - (1 - A\beta) p - \beta (A - B) \alpha \right| G_p(k, \lambda) a_k - (A - B) \beta (p - \alpha) \leq 0. \end{aligned}$$

Hence, by the principle of maximum modulus, it is asserted that  $f(z) \in S_p^*(A, B, \alpha, \beta, \lambda)$ .

Conversely, we suppose that  $f(z)$  is in the class  $S_p^*(A, B, \alpha, \beta, \lambda)$ , then by virtue of (12)

$$\left| \left\{ \frac{z (U_z^{(\lambda, p)} f(z))'}{U_z^{(\lambda, p)} f(z)} - p \right\} / \left\{ (p - \alpha) (A - B) + p B - B z \frac{(U_z^{(\lambda, p)} f(z))'}{U_z^{(\lambda, p)} f(z)} \right\} \right|$$

$$= \left| \frac{- \sum_{k=n+p}^{\infty} (k-p) G_p(k, \lambda) a_k z^k}{(A-B)(p-\alpha) z^p + \sum_{k=n+p}^{\infty} |B(k-p) - (A-B)(p-\alpha)| G_p(k, \lambda) a_k z^k} \right| < \beta. \quad (14)$$

Since  $|\operatorname{Re}(z)| \leq |z|$  for any  $z$ , we find from (14), that,

$$\operatorname{Re} \left[ \frac{\sum_{k=n+p}^{\infty} (k-p) G_p(k, \lambda) a_k z^k}{(A-B)(p-\alpha) z^p + \sum_{k=n+p}^{\infty} |B(k-p) - (A-B)(p-\alpha)| G_p(k, \lambda) a_k z^k} \right] < \beta.$$

Now choose values of  $z$  on the real axis, such that

$$\frac{z (U_z^{(\lambda, p)} f(z))'}{U_z^{(\lambda, p)} f(z)}$$

is real. Assuming  $z \rightarrow 1$ , through real values, we get

$$\sum_{k=n+p}^{\infty} (k-p) G_p(k, \lambda) a_k \leq (A-B) \beta(p-\alpha) + \sum_{k=n+p}^{\infty} \beta |B(k-p) - (A-B)(p-\alpha)| G_p(k, \lambda) a_k,$$

which asserts (13).

*Corollary 1 :* Let the function  $f(z)$ , defined by (2), belongs to the class  $S_p^*(A, B, \alpha, \beta, \lambda)$ , then

$$a_k \leq \frac{(A-B) \beta (p-\alpha)}{[(1-\beta B)(k-p) + \beta (A-B)(p-\alpha)] G_p(k, \lambda)}, \quad k \geq n+p. \quad (15)$$

*Remark 1* : Considering  $\lambda = 0$  and  $\lambda = 1$  respectively in Theorem 1, results due to Singh and Sohi<sup>8</sup> become particular cases of our investigation.

Similarly, by setting  $p = A = \beta = -B = 1$  in Theorem 1, result of Srivastava, Mishra and Das<sup>5</sup> become particular cases of our result.

### Distortion Theorem

*Theorem 2* : If  $f(z)$ , defined by (2), belongs to the class  $S_p^*(A, B, \alpha, \beta, \lambda)$ ,  $0 \leq \lambda < 1$ ; then for  $0 \leq \mu < 1$ , we have

$$|U_z^{(\mu, p)} f(z)| \geq |z|^p - \frac{(p+1)_n (A-B) \beta (p-\alpha)}{(p+1-\mu)_n G_p(n+p, \lambda) |(1-\beta B)n + \beta(A-B)(p-\alpha)|} |z|^{p+p} \quad (16)$$

and

$$|U_z^{(\mu, p)} f(z)| \leq |z|^p + \frac{(p+1)_n (A-B) \beta (p-\alpha)}{(p+1-\mu)_n G_p(n+p, \lambda) |(1-\beta B)n + \beta(A-B)(p-\alpha)|} |z|^{p+p} \quad (17)$$

for  $z \in \mathcal{D}$ . The estimates are sharp.

*Proof* : For  $0 \leq \lambda < 1$ ,

$$\left\{ \frac{\Gamma(k+1)}{\Gamma(k+1-\lambda)} \right\}_{k=n+p}^{\infty}$$

is an increasing sequence, and thus, from Theorem 1, we have

$$\begin{aligned} G_p(n+p, \lambda) \sum_{k=n+p}^{\infty} |(1-\beta B)(k-p) + \beta(A-B)(p-\alpha)| a_k \\ \leq \sum_{k=n+p}^{\infty} |(1-\beta B)(k-p) + \beta(A-B)(p-\alpha)| G_p(k, \lambda) a_k \leq (A-B) \beta (p-\alpha) \end{aligned}$$

$$\text{i.e. } \sum_{k=n+p}^{\infty} |(1 - \beta B)(k - p) + \beta(A - B)(p - \alpha)| a_k \leq \frac{(A - B)\beta(p - \alpha)}{G_p(n + p, \lambda)}, \quad (18)$$

which yields

$$\sum_{k=n+p}^{\infty} a_k \leq \frac{(A - B)\beta(p - \alpha)}{G_p(n + p, \lambda) |(1 - \beta B)n + (A - B)(p - \alpha)|}, \quad k \geq n + p. \quad (19)$$

Now, due to mutual adjustments of terms in (18) and (19), we may write the following :

$$\begin{aligned} \sum_{k=n+p}^{\infty} (1 - B\beta) k a_k &\leq \frac{(A - B)\beta(p - \alpha)}{G_p(n + p, \lambda)} |(1 - \beta B)p - \beta(A - B)(p - \alpha)| \sum_{k=n+p}^{\infty} a_k. \\ &\leq \frac{(A - B)\beta(p - \alpha)}{G_p(n + p, \lambda)} + \frac{|(1 - \beta B)p - \beta(A - B)(p - \alpha)| (A - B)\beta(p - \alpha)}{G_p(n + p, \lambda) |(1 - \beta B)n + \beta(A - B)(p - \alpha)|} \\ &= \frac{(A - B)\beta(p - \alpha)(1 - \beta B)(n + p)}{G_p(n + p, \lambda) |(1 - \beta B)n + \beta(A - B)(p - \alpha)|} \\ \text{i.e. } \sum_{k=n+p}^{\infty} k a_k &= \frac{(A - B)\beta(p - \alpha)(n + p)}{G_p(n + p, \lambda) |(1 - \beta B)n + \beta(A - B)(p - \alpha)|}. \end{aligned} \quad (20)$$

From (8), we have the expression, as follows :

$$U_z^{(\mu, p)} f(z) = z^p - \sum_{k=n+p}^{\infty} l_k z^k, \quad (21)$$

where

$$l_k = \frac{\Gamma(k + 1)\Gamma(p + 1 - \mu)}{\Gamma(p + 1)\Gamma(k + 1 - \mu)} a_k, \quad k \geq n + p. \quad (22)$$

We observe that, for  $0 \leq \mu < 1$ ,

$$\begin{aligned}
 l &\leq \frac{\Gamma(k+1)\Gamma(p+1-\mu)}{\Gamma(p+1)\Gamma(k+1-\mu)} \\
 &= \frac{\Gamma(p+1-\mu)}{\Gamma(k+1-\mu)} \frac{k}{(k-\mu)} \cdot \frac{(k-1)}{(k-\mu-1)} \cdots \frac{(1-n+p)}{(n+p-\mu+1)} \cdot \frac{\Gamma(n+p+1)}{\Gamma(n+p-\mu+1)} \\
 &\leq \frac{\Gamma(p+1-\mu)}{\Gamma(k+1-\mu)} \frac{k}{k-1} \cdot \frac{(k-1)}{(k-2)} \cdots \frac{(n+p+1)}{(n+p)} \cdot \frac{\Gamma(n+p+1)}{\Gamma(n+p-\mu+1)} \\
 &= \frac{\Gamma(p+1-\mu)}{\Gamma(p+1)\Gamma(n+p-\mu+1)} k. \tag{23}
 \end{aligned}$$

Now a mutual employment of (20) and (23), allows us to write

$$\begin{aligned}
 \sum_{k=n+p}^{\infty} l_k &\leq \frac{\Gamma(p+1-\mu)\Gamma(n+p)}{\Gamma(p+1)\Gamma(n+p-\mu+1)} \sum_{k=n+p}^{\infty} k a_k \\
 &= \frac{(p+1)_n (A-B) \beta (p-\alpha)}{(p+1-\mu)_n G_p(n+p, \lambda) \left| (1-\beta B) n + \beta (A-B) (p-\alpha) \right|}. \tag{24}
 \end{aligned}$$

Eventually

$$|U_z^{(\mu, p)} f(z)| \geq |z|^p - |z|^{n+p} \sum_{k=n+p}^{\infty} l_k$$

yields the results (16) due to (24). Similarly, (17) can be proved.

The sharpness of the theorem follows, if we consider

$$f(z) = z^p - \frac{(A-B) \beta (p-\alpha)}{G_p(n+p, \lambda) \left| (1-\beta B) n + \beta (A-B) (p-\alpha) \right|}. \tag{25}$$

*Corollary 2 :* By setting  $\mu = 0$  in Theorem 2, we obtain, for  $f(z) \in S_p^*(A, B, \alpha, \beta, \lambda)$ , the following :

$$|f(z)| \geq |z|^p - \frac{(A-B)\beta(p-\alpha)}{G_p(n+p, \lambda) |(1-\beta B)n + \beta(A-B)(p-\alpha)|} |z|^{n+p} \quad (26)$$

and

$$|f(z)| \leq |z|^p + \frac{(A-B)\beta(p-\alpha)}{G_p(n+p, \lambda) |(1-\beta B)n + \beta(A-B)(p-\alpha)|} |z|^{n+p}. \quad (27)$$

Estimates are, thus, sharp for  $f(z)$  given by (3.10).

*Corollary 3* : Assuming  $\mu \rightarrow 1$  and for  $f(z) \in S_p^*(A, B, \alpha, \beta, \lambda)$ , Theorem 2 reduces to

$$|f'(z)| \geq p |z|^p - \frac{(A-B)\beta(p-\alpha)(p+n)}{G_p(n+p, \lambda) |(1-\beta B)n + \beta(A-B)(p-\alpha)|} |z|^{n+p}, \quad (28)$$

and

$$|f'(z)| \leq p |z|^{p-1} - \frac{(A-B)\beta(p-\alpha)(p+n)}{G_p(n+p, \lambda) |(1-\beta B)n + \beta(A-B)(p-\alpha)|} |z|^{n+p-1}. \quad (29)$$

Sharpness of the estimation is verified as Cor. 3.

### Inclusion Theorem

*Theorem 3* : For  $0 \leq \lambda \leq \mu < 1$ ,  $0 \leq \alpha < p$ ,  $0 < \beta \leq 1$ ,  $-1 \leq B < A \leq 1$  and  $0 < A \leq 1$ , we have

$$S_p^*(A, B, \alpha, \beta, \mu) \subseteq S_p^*(A, B, \alpha, \beta, \lambda). \quad (30)$$

*Proof* : For  $f(z) \in S_p^*(A, B, \alpha, \beta, \mu)$ , Theorem 1 reduces to

$$\sum_{k=n+p}^{\infty} |(1-\beta B)(k-p) + \beta(A-B)(p-\alpha)| G_p(k, \mu) a_k \leq (A-B)\beta(p-\alpha), \quad (31)$$

where,  $G_p(k, \mu)$  may be expressed, by virtue of (9), as

$$G_p(k, \mu) = \frac{\Gamma(k+1) \Gamma(p+1-\mu)}{\Gamma(p+1) \Gamma(k+1-\mu)}.$$

For  $\mu$  to be restricted as above, and  $\phi(\mu)$  be assumed as an increasing function

$$\phi(\mu) = \frac{\Gamma(p+1-\mu)}{\Gamma(k+1-\mu)}$$

we observe that

$$\phi(\mu+1) - \phi(\mu) = \frac{\Gamma(p-\mu)}{\Gamma(k+1-\mu)} (k-p) > 0,$$

very clearly asserts that  $\phi(\mu)$  is definitely an increasing function of  $\lambda$  [since  $(k-p)>0$ ] and thus,

$$G_p(k, \lambda) \leq G_p(k, \mu). \quad (32)$$

Finally from (31), we have

$$\begin{aligned} & \sum_{k=n+p}^{\infty} |(1-\beta B)(k-p) + \beta(A-B)(p-\alpha)| G_p(k, \lambda) a_k \\ & \leq \sum_{k=n+p}^{\infty} |(1-\beta B)(k-p) + \beta(A-B)(p-\alpha)| G_p(k, \mu) a_k \\ & \leq (A-B) \beta (p-\alpha). \end{aligned}$$

This is an evidence, that,  $f(z) \in S_p^*(A, B, \alpha, \beta, \lambda)$ .

### Fractional Integral Operator

*Theorem 4 :* Let  $f(z) \in S_p^*(A, B, \alpha, \beta, \lambda)$ , be defined by (2) and  $c$  be real number, such that  $c > -p$ , then the function  $F(z)$ , define by

$$F(z) = \frac{c+p}{z^c} \int_0^z t^{c-1} f(t) dt, \quad (33)$$

also belongs to the above class.

*Proof :* By virtue of (33), it follows from (2) that,

$$F(z) = \frac{c+p}{z^c} \int_0^z t^{c-1} \left\{ t^p - \sum_{k=n+p}^{\infty} a_k t^k \right\} dt,$$

which upon integration and change of order of summation and integration, permissible under stated conditions, reduces to

$$F(z) = z^p - \sum_{k=n+p}^{\infty} b_k z^k, \quad (34)$$

where

$$b_k = \left( \frac{c+p}{c+k} \right) a_k. \quad (35)$$

Thus, we write

$$\begin{aligned} & \sum_{k=n+p}^{\infty} |(1 - \beta B)(k - p) + \beta (A - B)(p - \alpha)| G_p(k, \lambda) b_k \\ &= \sum_{k=n+p}^{\infty} |(1 - \beta B)(k - p) + \beta (A - B)(p - \alpha)| G_p(k, \lambda) \left( \frac{c+p}{c+k} \right) a_k \\ &\leq (A - B) \beta (p - \alpha). \end{aligned}$$

which is the justification, by virtue of Theorem 1, for  $F(z) \in S_p^*(A, B, \alpha, \beta, \lambda)$ , which completes the proof.

*Theorem 5* : Let  $c$  be a real number, such that  $c < -p$ , if  $F(z) \in S_p^*(A, B, \alpha, \beta, \lambda)$ , then the function  $f(z)$ , which is defined by (33), is  $p$ -valent in  $|z| < R_1$ , where

$$R_1 = \inf_{k \geq n+p} \left[ \frac{(c+p) \Gamma(k) \Gamma(p+1-\lambda) \{(1-\beta B)(k-p) + \beta(A-B)(p-\alpha)\}}{(c+k) \Gamma(p) \Gamma(k+1-\lambda) (A-B) \beta (p-\alpha)} \right]^{1/(k-p)} \quad (36)$$

and the result is sharp.

*Proof* : Let

$$F(z) = z^p - \sum_{k=n+p}^{\infty} a_k z^k,$$

it follows from (33), that,

$$\begin{aligned} f(z) &= z^{1-c} \frac{(z^c F(z))'}{c+p}, \\ \text{i.e.} \quad f(z) &= z^p - \sum_{k=n+p}^{\infty} \left( \frac{c+k}{c+p} \right) a_k z^k, \quad c > -p. \end{aligned} \quad (37)$$

To prove the required result, it is sufficient to prove that

$$\left| \frac{f'(z)}{z^{p-1}} - p \right| \leq p, \quad |z| < R_1. \quad (38)$$

Now,

$$\left| \frac{f'(z)}{z^{p-1}} - p \right| = \left| - \sum_{k=n+p}^{\infty} k \left( \frac{c+k}{c+p} \right) a_k z^{k-p} \right|,$$

thus, if

$$\left| \frac{f'(z)}{z^{p-1}} - p \right| \leq p ,$$

then

$$\sum_{k=n+p}^{\infty} \frac{k}{p} \left( \frac{c+k}{c+p} \right) a_k |z|^{k-p} \leq 1 . \quad (39)$$

Also, since  $F(z) \in S_p^*(A, B, \alpha, \beta, \lambda)$ , so by Theorem 1, we have

$$\sum_{k=n+p}^{\infty} \frac{|(1-\beta B)(k-p) + \beta(A-B)(p-\alpha)| G_p(k, \lambda) a_k}{(A-B)\beta(p-\alpha)} \leq 1 . \quad (40)$$

Hence (39) will be satisfied, if

$$\frac{k}{p} \left( \frac{c+k}{c+p} \right) |z|^{k-p} \leq \frac{|(1-\beta B)(k-p) + \beta(A-B)(p-\alpha)| G_p(k, \lambda)}{(A-B)\beta(p-\alpha)}$$

i.e.  $|z| \leq \left[ \frac{(c+p)\Gamma(k)\Gamma(p+1-\lambda)}{(c+k)\Gamma(p)\Gamma(k+1-\lambda)} \frac{|(1-\beta B)(k-p) + \beta(A-B)(p-\alpha)|}{(A-B)\beta(p-\alpha)} \right]^{1/(k-p)} , k \geq n+p . \quad (41)$

Therefore, this proves that  $f(z)$  is  $p$ -valent in  $|z| < R_1$ . Sharpness follows, if we consider

$$f(z) = z^p - \frac{(c+k)(A-B)\beta(p-\alpha)}{(c+p)|(1-\beta B)(k-p) + \beta(A-B)(p-\alpha)| G_p(k, \lambda)} , k \geq n+p . \quad (42)$$

### Radii of Starlikeness and Convexity

*Theorem 6 :* The function  $f(z)$ , defined by (2) be in the class  $S_p^*(A, B, \alpha, \beta, \lambda)$ , then  $f(z) \in S^*(p, \delta)$ ,  $0 \leq \delta < p$  in  $|z| < R_2$ , where

$$R_2 = \inf_{k \geq n+p} \left[ \frac{(p-\delta) \left\{ (1-\beta B) (k-p) + \beta(A-B) (p-\alpha) \right\}}{(k-\delta) (A-B) \beta (p-\alpha)} \right]^{1/(k-p)} \quad (43)$$

The result is sharp.

*Proof:* In order to establish the required result, we deem it sufficient to prove that

$$\left| \frac{zf'(z)}{f(z)} - p \right| \leq p - \delta, \quad |z| \leq R_2. \quad (44)$$

Now

$$\begin{aligned} \left| \frac{zf'(z)}{f(z)} - p \right| &= \left| \frac{\sum_{k=n+p}^{\infty} (k-p) a_k z^k}{z^p - \sum_{k=n+p}^{\infty} a_k z^k} \right|, \\ &\leq \frac{\sum_{k=n+p}^{\infty} (k-p) a_k |z|^{k-p}}{1 - \sum_{k=n+p}^{\infty} a_k |z|^{k-p}}, \end{aligned}$$

thus if,  $\left| \frac{zf'(z)}{f(z)} - p \right| \leq p - \delta,$

then, it is obvious to write

$$\sum_{k=n+p}^{\infty} \left( \frac{k-\delta}{p-\delta} \right) a_k z^{k-p} \leq 1. \quad (45)$$

Also, as  $f(z) \in S_p^*(A, B, \alpha, \beta, \lambda)$ , by virtue of Theorem 1, we write

$$\sum_{k=n+p}^{\infty} \frac{|(1 - \beta B)(k-p) + \beta(A-B)(p-\alpha)| G_p(k, \lambda)}{(A-B)\beta(p-\alpha)} \leq 1.$$

Therefore, (45) is justified, if

$$|z| \leq \left[ \frac{(p-\delta)|(1 - \beta B)(k-p) + \beta(A-B)(p-\alpha)|}{(k-\delta)(A-B)\beta(p-\alpha)} G_p(k, \lambda) \right]^{1/(k-p)}. \quad (46)$$

This concludes that,  $f(z) \in S^*(p, \delta)$ , i.e.  $f(z)$  is  $p$ -valently starlike of order  $\delta$  in  $|z| < R_2$ . Sharpness follows from the function defined as

$$f(z) = z^p - \frac{(A-B)\beta(p-\alpha)}{|(1 - \beta B)(k-p) + \beta(A-B)(p-\alpha)| G_p(k, \lambda)} z^k. \quad (47)$$

*Theorem 7 :* Let the function  $f(z)$ , defined by (2), belongs to  $S_p^*(A, B, \alpha, \beta, \lambda)$ , then  $f(z) \in C(p, \delta)$ ,  $0 \leq \delta < p$  in the disc  $|z| < R_3$ , where

$$R_3 = \inf_{k \geq n+p} \left[ \frac{p(p-\delta)|(1 - \beta B)(k-p) + \beta(A-B)(p-\alpha)|}{k(k-\delta)(A-B)\beta(p-\alpha)} G_p(k, \lambda) \right]^{1/(k-p)} \quad (48)$$

The result is sharp.

*Proof :* To prove the statement completely, it is sufficient to show that

$$\left| \left\{ 1 + \frac{zf''(z)}{f'(z)} \right\} - p \right| \leq (p-\delta), \quad |z| \leq R_3.$$

$$\text{we have,} \quad \left| \left\{ 1 + \frac{zf''(z)}{f'(z)} \right\} - p \right| = \left| \frac{\sum_{k=n+p}^{\infty} k(k-p) a_k z^{k-p}}{p - \sum_{k=n+p}^{\infty} k a_k z^{k-p}} \right|$$

$$\leq \frac{\sum_{k=n+p}^{\infty} k(k-p) a_k |z|^{k-p}}{p - \sum_{k=n+p}^{\infty} k a_k |z|^{k-p}}. \quad (50)$$

Therefore, if (49) is true, then

$$\sum_{k=n+p}^{\infty} k(k-p) a_k |z|^{k-p} \leq (p-\delta) \left\{ p - \sum_{k=n+p}^{\infty} k a_k |z|^{k-p} \right\}$$

i.e.  $\sum_{k=n+p}^{\infty} \frac{k}{p} \cdot \frac{(k-\delta)}{(p-\delta)} a_k |z|^{k-p} \leq 1. \quad (51)$

By virtue of Theorem 1, (13) holds true, and thus (51) will be justified if

$$|z| \leq \left[ \frac{p(p-\delta) |(1-\beta B)(k-p) + \beta(A-B)(p-\alpha)| G_p(k, \lambda)}{k(k-\delta)(A-B)\beta(p-\alpha)} \right]^{1/(k-p)}.$$

Thus  $f(z) \in C(p, \delta)$ , i.e.  $f(z)$  is  $p$ -valently convex of order  $\delta$  in  $|z| < R_3$ , while sharpness for  $f(z)$  follows from (47). The theorem is thereby asserted.

### Acknowledgements

This work is supported by CSIR Research Scheme No. 25 (0105)/99/EMR-II, sanctioned to first author (PKB). The second author (GMS) is thankful to Al-Azhar University of Gaza, Palestine, for financial assistance.

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# On the equation for electron – proton pairing at finite temperature

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Received July 22, 2000; Accepted April 10, 2001

## Abstract

An equation was considered recently to describe electron-proton pairing at finite temperature. It was shown to have a solution in terms of a difference equation which, on numerical evaluation, led to a temperature-dependent bound state spectrum. In the present paper, the solution of the pairing equation is discussed afresh, resulting in an independent (and elementary) derivation of the crucial difference equation.

(Keywords : finite temperature theory/electron proton pairing/x-ray astronomy)

## Introduction

Some time back, an equation was obtained<sup>1</sup> which described the pairing of an electron and a proton at finite temperature. Under a suitable approximation, this equation yielded a bound state spectrum in the temperature regime around one million degrees Kelvin. The deepest of the binding energies predicted in this spectrum were in the KeV range. This meant that the bound states could actually sustain themselves in the system, despite its high temperature, since the background thermal energy per particle, which would only be in the range of few hundred eV, would not be enough to break up the former.

The above finding may be quite relevant to an understanding of Solar coronal active regions and solar flares. In particular, the mechanism for the massive energy production associated with these phenomena could be provided by falling into the paired state by the electrons and the protons of the astrophysical plasma. Furthermore, the emission spectrum of the paired state in certain possible stable or quasi-stable configurations corresponding to definite temperatures, could also explain a large part of the X-ray emission line data from the flares and the other active regions of the Sun and its corona. Preliminary work along these lines reported in the reference quoted above and in later papers<sup>2,3</sup> has led to encouraging results.

Of course, the occurrence of sustainable bound states at high temperatures is a rather unexpected result. So despite its good consequences, the basis of this result needs detailed probing. Some work in this direction has been published<sup>4</sup>. More recently the author<sup>5</sup> has given a systematic derivation of a coupled set of equations for electron-proton pairing at finite temperature and shown that in the static Coulomb limit they indeed lead to the pairing equation used by us earlier. In the present paper, we try a new approach to solving the approximate version of the pairing equation. This is done by transforming the equation onto coordinate space where the ansatz for the solution can be taken as a sum over the familiar Laguerre polynomials. The coefficients in the sum are then found to obey a difference equation the solution of which leads to the bound state spectrum. This difference equation is identical to the one obtained earlier from momentum-space methods using the less familiar four-dimensional spherical harmonics given in terms of Gegenbauer polynomials.

### The Pairing Equation in Coordinate Space

The pairing equation in momentum space in the static Coulomb limit is given by

$$\left( W - \frac{p^2}{2\mu} \right) \psi (\vec{p}) = \frac{\alpha}{4\pi^2} Q(w, p) \int \frac{d\vec{q}}{(\vec{q} - \vec{p})^2} \psi (\vec{q}), \quad (1)$$

$$Q(w, p) = \tan h \left[ (\mu_a w - \epsilon_p^a) \frac{\beta}{2} \right], \tan h \left[ (\mu_b w - \epsilon_p^b) \frac{\beta}{2} \right] \quad (2)$$

where  $w = -|w|$  stands for the pair binding energy,  $\vec{p}$  or  $\vec{q}$  for the (relative) pair momentum;  $\alpha = (e^2 / 4\pi)$  is the fine structure constant in natural units ( $(\hbar = c = 1)$ ) which we use throughout;  $a$  and  $b$  stand, respectively, for the electron and the proton and

$$\mu = \frac{m_a m_b}{m_a + m_b}, \quad \mu_{a, b} = \frac{m_{a, b}}{m_a + m_b} \quad (3)$$

$$\epsilon_p^{a, b} = \frac{p^2}{2m_{a, b}}, \quad \beta = \frac{1}{KT}$$

where  $k$  is the Boltzmann constant and  $T$  the temperature.

It may be noted that

$$Q(w, p) = \frac{a - \tan h(\beta p^2 / 4m_a)}{1 - a \tan h(\beta p^2 / 4m_a)} - \frac{b - \tan h(\beta p^2 / 4m_b)}{1 - b \tan h(\beta p^2 / 4m_b)} \quad (4)$$

where

$$a = \tan h(\beta \mu_a w / 2), \quad b = \tan h(\beta \mu_b w / 2). \quad (5)$$

We now make the approximation

$$\tan h\left(\frac{\beta p^2}{4m_{a,b}}\right) = \frac{\beta p^2}{4m_{a,b}} \quad (6)$$

and combine the two terms in eqn. (4), dropping terms of order  $p^4$  and higher in both the numerator and the denominator in keeping with the spirit of (6). So we get

$$Q(w, p) = \frac{d_1 - d_2 p^2}{1 - d_3 p^2} \equiv \tilde{Q}(w, p) \quad (7)$$

where

$$d_1 = a + b, \quad d_2 = \beta(1 + ab) / 4\mu, \quad d_3 = \frac{\beta}{4} \left( \frac{a}{m_a} + \frac{b}{m_b} \right) \quad (8)$$

Next, we replace  $Q$  in eqn. (1) by  $\tilde{Q}$  and write the resulting approximate version of the equation as

$$(1 - d_3 p^2)(w - p^2/2\mu) \psi(\vec{p}) = \frac{\alpha}{4\pi^2} (d_1 - d_2 p^2) \int \frac{d\vec{q}}{(\vec{q} - \vec{p})^2} \psi(\vec{q}) \quad (9)$$

On multiplying both sides by

$$\frac{1}{(2\pi)^{3/2}} \int d\vec{p} e^{-\vec{p} \cdot \vec{r}}$$

and going through the standard manipulations, we can transform this equation onto coordinate space to get

$$(1 + d_3 \nabla^2) \left( W + \frac{\nabla^2}{2\mu} \right) \psi(\vec{r}) = \frac{\alpha}{2} \left( \frac{d_1}{r} \psi(\vec{r}) + d_2 \nabla^2 \left[ \frac{\psi(\vec{r})}{r} \right] \right) \quad (10)$$

where

$$\psi(\vec{r}) = \frac{1}{(2\pi)^{3/2}} \int d\vec{p} e^{-i\vec{p}\cdot\vec{r}} \psi(\vec{p}). \quad (11)$$

### Solution of the Pairing Equation in Coordinate Space

We observe that in the absence of the terms containing  $d_2$  and  $d_3$ , the pairing equation in coordinate space, namely, eqn. (10) above, has the same structure as the Schrodinger equation for the hydrogen atom. The solution in that case is :

$$\psi(\vec{r}) = R(r) Y_l^m(\theta, \phi) = \frac{u(r)}{r} Y_l^m(\theta, \phi) \quad (12)$$

$$u(r) = u_{m,l}(r) = N' r^{l+1} e^{-p_0 r} L_{n-l-1}^{2l+1}(2p_0 r) \quad (13)$$

where  $Y_l^m(\theta, \phi)$  are the three dimensional spherical harmonics with  $l$  and  $m$  standing, respectively, for the angular momentum and the azimuthal quantum numbers ( $l = 0, 1, 2, \dots$ ;  $m$  for a given  $l$  takes the  $2l+1$  values  $-l, -l+1, \dots, l-1, l$ ); the  $L$ 's denote the Laguerre polynomials in which the index  $n$ , the principal quantum number, takes on the values  $l+1, l+2, \dots$ , i.e., for any given  $l, n \geq l+1$ ;  $N'$  is the normalisation constant.

Now, as regards eqn. (10), we can once more try separation of variables, and choose  $\psi = RY = (u/r) Y$  with unknown  $R$  (or  $u$ ) and  $Y$ . The spherical symmetry of the problem enables us to choose  $Y_l^m$  for  $Y$ . So we can still use eqn. (12). We can then also write (see for instance Ch. 7 in Powell and Crasemann<sup>6</sup>).

$$\begin{aligned} \nabla^2 &= \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial}{\partial r} \right) + \frac{1}{r^2} \left[ \frac{1}{\sin\theta} \frac{\partial}{\partial\theta} \left( \sin\theta \frac{\partial}{\partial\theta} \right) + \frac{1}{\sin^2\theta} \frac{\partial^2}{\partial\phi^2} \right] \\ &= \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial}{\partial r} \right) - \frac{l(l+1)}{r^2}. \end{aligned} \quad (14)$$

With the use of eqn. (12) and (14), eqn. (10) takes the form

$$\begin{aligned}
 & (1 + d_3 \nabla^2) \left( -p_0^2 \frac{u}{r} Y_l^m + \frac{1}{r} \frac{d^2 u}{dr^2} Y_l^m - \frac{l(l+m)}{r^2} \frac{u}{r} Y_l^m \right) \\
 & = \alpha \mu (d_1 + d_2 \nabla^2) \frac{u}{r^2} Y_l^m
 \end{aligned} \tag{15}$$

where we have put

$$p_0^2 = -2\mu w = 2\mu |w|. \tag{16}$$

It is more convenient to work with the dimensionless variable

$$y = 2p_0 r \tag{17}$$

and

$$\nabla_y^2 = (1/4 p_0^2) \nabla^2 \tag{18}$$

in terms of which eqn. (15) is given by

$$\begin{aligned}
 & \left( 1 + 4p_0^2 d_3 \nabla_y^2 \right) \left( \frac{-u}{4y} - \frac{l(l+1)}{y^3} u + \frac{1}{y} \frac{d^2 u}{dy^2} \right) Y_l^m \\
 & = \frac{\alpha \mu}{2p_0} (d_1 + 4p_0^2 d_2 \nabla_y^2) \frac{u}{y^2} Y_l^m.
 \end{aligned} \tag{19}$$

To guess a suitable form for  $u$ , we observe that for a given  $\alpha (= 2l + 1)$ , the Laguerre polynomials  $L_k^\alpha(y)$ , where  $k (= n - l - 1) = 0, 1, 2, \dots$ , form a complete set of orthogonal functions in  $y$  space ( $0 < y < \infty$ ) with respect to the weight functions  $e^{-y} y^{(\alpha+l)}$ . So  $u(y)$  can certainly be written as

$$u(y) = u_l(y) = N e^{-y/2} y^{(\alpha+1)/2} \sum_{n \geq l+1}^{\infty} b_n L_{n-l-1}^\alpha(y) \tag{20}$$

where the coefficients  $b_n$  are as such arbitrary and  $N$  is just the usual normalisation constant. It is preferable to use, in place of  $b_n$ , the coefficients  $a_{n-l}/n^2$ , where the denominator  $n^2$  is

chosen for later convenience and the choice of the index  $n - 1$  on  $\alpha$  is useful because for a given  $l$ , the first coefficient in the series is simply  $a_l$  (to be followed by  $a_{l+1}, a_{l+2}, \dots$ ) and we can thus easily keep track of the particular angular momentum value we are dealing with. So we have

$$u_l(y) = Ne^{-y/2} y^{(\alpha+1)/2} \sum_{n \geq l+1}^{\infty} \frac{a_{n-1}}{n^2} L_{n-l-1}^{\alpha}(y). \quad (20a)$$

This may also be written as

$$\begin{aligned} u_l(y) &= Ne^{-y/2} y^{(\alpha+1)/2} \sum_{k=0}^{\infty} \frac{a_{k+l}}{(k+l+1)^2} L_k^{\alpha}(y) \\ &= V_l \sum_{k=0}^{\infty} \hat{L}_k^{\alpha}(y) \end{aligned} \quad (20b)$$

where

$$\alpha = 2l+1, \quad k = n - l - 1 \quad (21)$$

and we have put

$$\frac{a_{k+l}}{(k+l+1)^2} L_k^{\alpha}(y) = \hat{L}_k^{\alpha}, \quad Ne^{-y/2} y^{(\alpha+1)/2} = V_l. \quad (22)$$

On substituting the above expression for  $u$  in eqn. (19), and using the equation followed by the Laguerre polynomials, namely,

$$y \frac{d^2 L_k^{\alpha}(y)}{dy^2} + (\alpha + 1 - y) \frac{dL_k^{\alpha}(y)}{dy} + kL_k^{\alpha}(y) = 0, \quad (23)$$

we get  $q(1 + 4p_0^2 d_3 \nabla_y^2) \frac{V_l \hat{L}_k^{\alpha}}{y^2} Y_l^m = \frac{\lambda}{2p_0} (d_1 + 4p_0^2 d_2 \nabla_y^2) \frac{V_l \hat{L}_k^{\alpha}}{y^2} Y_l^m$  (24)

where  $\lambda = -\alpha \mu, q = k + l + 1$ . (25)

The sum over  $k$  in eqn. (24) has been suppressed for the present and will be put in later. It can easily be checked that

$$\nabla_y^2 \frac{V_l \hat{L}_k^\alpha}{y^2} Y_l^m = \left[ \frac{1}{4y^2} - \frac{(k+l)}{y^3} - \frac{2l}{y^4} \right] V_l \hat{L}_k^\alpha Y_l^m - \frac{2}{y^3} V_l \frac{d\hat{L}_k^\alpha}{dy} Y_l^m. \quad (26)$$

With the help of the standard recurrence relations

$$y \frac{dL_k^\alpha}{dy} = k L_k^\alpha - (k + 2l + 1) L_{k-1}^\alpha, \quad (27)$$

$$y \frac{dL_k^\alpha}{dy} = (k+l) L_{k+l}^\alpha - (k+2l+2-y) L_k^\alpha, \quad (28)$$

the last term in eqn. (26) can be written as

$$\frac{-2}{y^4} y \frac{d\hat{L}_k^\alpha}{dy} V_l Y_l^m = \left[ \frac{2l+2}{y^4} \hat{L}_k^\alpha + \frac{(k+2l+1)}{y^4} \hat{L}_{k-1}^\alpha - \frac{(k+1)}{y^4} \hat{L}_{k+l}^\alpha - \frac{1}{y^3} \hat{L}_k^\alpha \right] V_l Y_l^m \quad (29)$$

$$\text{where } \hat{L}_{k\pm i}^\alpha = \frac{a_{k+l}}{(k+l+1)^2} L_{k\pm i}^\alpha; \quad i = 0 \pm 1, \pm 2, \dots \quad (30)$$

So we get

$$\begin{aligned} \nabla_y^2 \frac{V_l \hat{L}_k^\alpha}{y^2} Y_l^m &= \left[ \frac{1}{4y^2} \hat{L}_k^\alpha - \frac{(k+l+1)}{y^3} \hat{L}_k^\alpha + \frac{2}{y^4} \hat{L}_k^\alpha + \frac{(k+2l+1)}{y^4} \hat{L}_{k-1}^\alpha - \frac{(k+1)}{y^4} \hat{L}_{k+l}^\alpha \right] V_l Y_l^m \\ &= \left[ \frac{1}{4y^2} \hat{L}_k^\alpha + A_{k,l} \right] V_l Y_l^m. \end{aligned} \quad (31)$$

On substituting this result in eqn. (24), we get

$$(1 + p_0^2 d_3)q \frac{V_l \hat{L}_k^\alpha}{y^2} Y_l^m + 4p_0^2 d_3 q A_{k,l} V_l Y_l^m$$

$$= \frac{\lambda}{2p_0} (d_1 + p_0^2 d_2) \frac{V_l \hat{L}_k^\alpha}{y^2} Y_l^m + \frac{\lambda}{2p_0} 4p_0^2 d_2 A_{k,l} V_l Y_l^m. \quad (32)$$

On multiplying throughout by  $y^2$ , dropping the common factor  $V_l Y_l^m$ , and using

$$\begin{aligned} C_1 &= 1 - d_3 p_0^2, \quad C_2 = (\lambda/2p_0) (d_1 - d_2 p_0^2) \\ C_3 &= 1 + d_3 p_0^2, \quad C_4 = (\lambda/2p_0) (d_1 + d_2 p_0^2) \end{aligned} \quad (33)$$

$$\text{so that} \quad p_0^2 d_3 = (C_3 - C_1)/2, \quad (\lambda/2 p_0) d_2 p_0^2 = (C_4 - C_2)/2, \quad (34)$$

eqn. (32) can be put in the form

$$\begin{aligned} (qC_3 - C_4) y^2 \hat{L}_k^\alpha + 2[q(C_3 - C_1) - (C_4 - C_2)] [(q+l) \hat{L}_{k-1}^\alpha + 2\hat{L}_k^\alpha \\ - (q-l) \hat{L}_{k+1}^\alpha - qy \hat{L}_k^\alpha]. \end{aligned} \quad (35)$$

The factors  $y$  and  $y^2$  can now be removed by the application of the recurrence relation

$$y \hat{L}_k^\alpha (y) = 2q \hat{L}_k^\alpha (y) - (q-l) \hat{L}_{k+1}^\alpha (y) - (q+l) \hat{L}_{k-1}^\alpha (y). \quad (36)$$

Eqn. (35) then takes the form

$$A(q, l) \hat{L}_{k+2}^\alpha + B(q, l) \hat{L}_{k+1}^\alpha + C(q, l) \hat{L}_k^\alpha + D(q, l) \hat{L}_{k-1}^\alpha + E(q, l) \hat{L}_{k-2}^\alpha = 0 \quad (37)$$

where

$$\begin{aligned} A(q, l) &= [qC_3 - C_4] [q-l] [q-l+1] \\ B(q, l) &= [qC_3 - C_4] [(-2q)(q-l) - 2(q-l)(q+1)] \\ &\quad + 2[q(C_3 - C_1) - (C_4 - C_2)] [q(q-l) - (q-l)] \\ C(q, l) &= [q(C_3 - C_4)] [4q^2 + (q-l)(q+l+1) + (q+l)(q-l-1)] \\ &\quad + 2[q(C_3 - C_1) - (C_4 - C_2)] [-2q^2 + 2] \end{aligned} \quad (38)$$

$$\begin{aligned}
 D(q, l) &= [q C_3 - C_4] [-2q(q+l) - 2(q-1)(q+l)] \\
 &\quad + 2[q(C_3 - C_1) - (C_4 - C_2)] [q(q+l) + (q+l)] \\
 E(q, l) &= [q C_3 - C_4] [q+l] [q+l-1].
 \end{aligned}$$

Adding and substituting  $2(q-l)$ ,  $4$  and  $2(q+l)$ , respectively, in the second brackets of the first terms of  $B(q, l)$ ,  $C(q, l)$  and  $D(q, l)$ , and doing the cancellations that follow, we get,

$$\begin{aligned}
 B(q, l) &= [q C_3 - C_4] [-2(q-l)(q+2)] - 2[q C_1 - C_2][(q-l)(q-1)] \\
 &= B_1(q, l) + B_2(q, l) \\
 C(q, l) &= [q C_3 - C_4] [2(q-l)(q+l) - 2(l-2)] \\
 &\quad - 2[q C_1 - C_2] [-2(q+1)(q-1)] \\
 &= [q C_3 - C_4] [(q-l)(q+2)(q+l+1)]/q \\
 &\quad + [q C_3 - C_4] [(q-l-1)(q-2)(q+l)]/q \\
 &\quad - 2[q C_1 - C_2] [-2(q+1)(q-1)] \\
 &= C_1(q, l) + C_2(q, l) + C_3(q, l) \\
 D(q, l) &= [q C_3 - C_4] [-2(q+l)(q-2)] - 2[q C_1 - C_2][(q+l)(q+1)] \\
 &= D_1(q, l) + D_2(q, l) \tag{39}
 \end{aligned}$$

where, in the breaking up of  $C(q, l)$ , we have used

$$2(q-l)(q+l) - 2(l-2) = (q+l+1)(q+2)(q-l)/q + (q+l)(q-2)(q-l-1)/q. \tag{40}$$

Eqn. (37) can thus be written as

$$\hat{L}_{k-2}^\alpha E(q, l) + \hat{L}_{k-1}^\alpha D_2(q, l) + \hat{L}_k^\alpha C_2(q, l) +$$

$$+ \hat{L}_{k-1}^\alpha D_1(q, l) + \hat{L}_k^\alpha C_3(q, l) + \hat{L}_{k+1}^\alpha B_1(q, l) \quad (41)$$

$$+ \hat{L}_k^\alpha C_1(q, l) + \hat{L}_{k+1}^\alpha B_2(q, l) + \hat{L}_{k+2}^\alpha A(q, l) = 0.$$

Let us now put in the sum over  $k$  which had been suppressed so far for notational convenience. We also note that

$$q = k + l + 1$$

$$\hat{L}_{k\pm i}^\alpha = \frac{a_{k+l}}{(k+l+1)^2} \hat{L}_{k\pm i}^\alpha; \quad i = 0, \pm 1, \pm 2 \dots$$

Thus the last term in eqn. (41) can, for instance, be written as

$$\begin{aligned} \sum_{k=0}^{\infty} \hat{L}_{k+2}^\alpha A(q, l) &= \sum_{k=0}^{\infty} \frac{a_{k+l}}{(k+l+1)^2} [qC_3 - C_4] [q-l] [q-l+1] L_{k+2}^\alpha \\ &= \sum_{q=l+1}^{\infty} \frac{a_{q-1}}{q^2} [qC_3 - C_4] [q-l] [q-l+1] L_{q-l+1}^\alpha \\ &= \sum_{q=l+3}^{\infty} [q=l-1] \frac{[q-2]}{[q-1]} \left[ \left( C_3 - \frac{C_4}{[q-2]} \right) \frac{[q-l-2][q-1]}{[q-2]^2} a_{q-3} \right] L_{q-l-1}^\alpha \quad (42) \end{aligned}$$

Expressing all the sums in eqn. (41) in terms of  $L_{q-l-1}^\alpha$  in a similar way, and remembering that  $L_{q-l-1}^\alpha \neq 0$  only for  $q \geq l+1$ , we get

$$\begin{aligned} &\sum_{q \geq l+1} \frac{[q+l+1][q+2]}{[q+1]} \left\{ \left[ C_3 - \frac{C_4}{[q+2]} \right] \frac{[q+l+2][q+1]}{[q+2]^2} a_{q+1} \right. \\ &\quad \left. - 2 \left[ C_1 - \frac{C_2}{[q+1]} \right] a_q + \left[ C_3 - \frac{C_4}{q} \right] \frac{[q-l][q+1]}{q^2} a_{q-1} \right\} L_{q-l-1}^\alpha \\ &- 2 \sum_{q \geq l+1} \frac{[q+1][q-1]}{q} \left\{ \left[ C_3 - \frac{C_4}{[q+1]} \right] \frac{[q+l+1]q}{[q+1]^2} a_q - 2 \left[ C_1 - \frac{C_2}{q} \right] a_{q-1} \right\} \end{aligned}$$

$$\begin{aligned}
& + \left[ C_3 - \frac{C_4}{[q-1]} \right] \frac{[q-l-1]q}{[q-1]^2} a_{q-2} \right\} L_{q-l-1}^\alpha \\
& + \sum_{q \geq l+2} \frac{[q-l-1][q-2]}{[q-1]} \left\{ \left[ C_3 - \frac{C_4}{q} \right] \frac{[q+l][q-1]}{[q]^2} a_{q-1} \right. \\
& \left. - 2 \left[ c_1 - \frac{c_2}{[q-1]} \right] a_{q-2} + \left[ c_3 - \frac{c_4}{[q-2]} \right] \frac{[q-l-2][q-1]}{[q-2]^2} a_{q-3} \right\} L_{q-l-1}^\alpha = 0. \quad (43)
\end{aligned}$$

It can be easily checked now that the solution of this equation is the simple difference equation

$$\left[ c_3 - \frac{c_4}{[q+1]} \right] \frac{[q+l+1]}{[q+l]^2} q a_q - 2 \left[ c_1 - \frac{c_2}{q} \right] a_{q-1} + \left[ c_3 - \frac{c_4}{[q-1]} \right] \frac{[q-l-1]q}{[q-1]^2} a_{q-2} = 0;$$

$$q \geq l+1, a_{l-1} = 0$$

or, equivalently

$$\left[ c_3 - \frac{c_4}{[q+2]} \right] \frac{[q+l+2][q+1]}{[q+2]^2} a_{q+1} - 2 \left[ c_1 - \frac{c_2}{[q+1]} \right] a_q + \left[ c_3 - \frac{c_4}{q} \right] \frac{[q-l][q+1]}{q^2} a_{q-1} = 0; \quad (44b)$$

$$q \geq l, a_{l-1} = 0.$$

If we define a new set of coefficients  $b_q$  by

$$b_q = a_q \frac{[q+l+1]}{[q+1]^3} [(q+1) c_3 - c_4], \quad (45)$$

eqn. (44b) can be transformed to

$$b_{q+1} - b_q \frac{2[q+1]}{[q+l+1]} \frac{[(q+1)c_1 - c_2]}{[(q+1)c_3 - c_4]} + b_{q-1} \frac{[q-l]}{[q+l]} = 0; \quad q \geq l, \quad b_{l-1} = 0 \quad (46)$$

his last form is convenient for computer calculations.

Eqn. (46) is of course identical to the difference equation which led us earlier to the bound state spectrum at high temperature<sup>1</sup>. It is hoped that the elementary derivation of the equation presented here will be useful to anyone interested in exploring the bound state spectra at finite temperature.

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## Synthesis and antimicrobial activity of some new chalkones

Received July 14, 2000; Revised June 15, 2001; Accepted August 25, 2001

### Abstract

2'-Hydroxy-1-(substituted phenyl)-3-(4-bromo-1-naphthyl)-2-propen-1-one were prepared by employing Claisen-Schmidt condensation. Structures of these compounds were confirmed by IR, PMR spectra and elemental analysis. All these compounds were found to be antibacterial and antifungal.

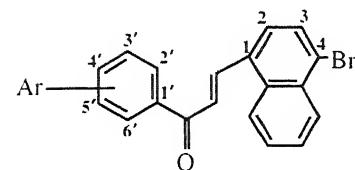
(Keywords : IR/PMR/elemental analysis/chalkones/antimicrobial agents)

Chalkones display interesting biological activities, including bactericidal<sup>1-5</sup>, fungicidal<sup>6</sup>, antimalarial<sup>7</sup>, antiinflammatory<sup>8</sup>, cytotoxic<sup>9</sup>, anticancer<sup>10</sup> and anti-invasive<sup>11</sup> properties. Hydroxychalkones have been widely investigated due to their synthetic potential and biological activity. In our continuing study of biologically active heterocyclic compounds like flavanones, flavones, flavonols, 2-pyrazoline and 1, 5-benzothiazepines, we discovered that the hydroxychalkones which serve as precursors of these heterocycles displayed potent antibacterial and antifungal activities against several human and plant pathogens. However, 2' or 4'-hydroxychalkones possessing naphthyl ring have not received considerable attention. Hence, we are reporting some new chalkones synthesised by a base-catalysed Claisen-Schmidt condensation of appropriately substituted 2' or 4'-hydroxyacetophenone and 4-bromo-1-naphthaldehyde.

2'-Hydroxy substituted acetophenone on treating with 4-bromo-1-naphthaldehyde in presence of ethanolic potassium hydroxide for 16h and on acidification with hydrochloric acid gave respective chalkone. They gave violet red colouration with ferric chloride and positive Wilson test<sup>12</sup>. Infrared spectra of these compounds in nujol mull showed a band in the region 1620-1650  $\text{cm}^{-1}$  due to carbonyl group and a band between 1610-1630  $\text{cm}^{-1}$  due to ethylenic double bond. These assignments are in agreement with those observed by Mohanty *et al.*<sup>13</sup>, Dhar and Gupta<sup>14</sup> and Vibhute and Jagdale<sup>15</sup>.

PMR spectra of a few representative chalkones were recorded in  $\text{CDCl}_3$  on Varian T-60 spectrometer using TMS as an internal reference. Compound-2 (Table 1) showed singlet at  $\delta$ 2.05 due to Ar-CH<sub>3</sub> and multiplet at  $\delta$ 6.9 to  $\delta$ 8.77 due to aromatic and ethylenic protons and one singlet at  $\delta$ 12.8 due to a phenolic proton. These observations are in agreement with those observed by Cardillo *et al.*<sup>16</sup>

Table I – Analytical data and antimicrobial activity of chalkones,



Com No.	Chalkones Ar	Elemental analysis		Antibacterial activity		Antifungal activity			
		C, H & Obsd. (Calcd.)	X	Diameter of Zone of Inhibition in mm		Germination (%) after 12 h			
				<i>E. coli</i>	<i>S. aureus</i>	<i>C. lunata</i>	<i>H. oryzae</i>		
						% ger- mina- tion	length of germ tube (μ)	% ger- mina- tion	length of germ tube (μ)
1.	2'-OH	C 64.76 (64.59) H 3.82 (3.68)	22.24 (22.66)	—	—	100	400	100	180
2.	2'-OH, 5'-CH <sub>3</sub>	C 65.10 (65.40) H 3.85 (4.09)	21.67 (21.80)	16	—	100	380	100	190
3.	2'-OH, 5'-Cl		29.40 (29.81)	—	—	100	320	100	240
4.	2'-OH, 5'-Br		36.70 (37.04)	42	33	30	48	90	352
5.	2'-OH, 4'CH <sub>3</sub> , 5'-Cl		29.25 (29.50)	18	10	95	310	80	280
6.	2'-OH, 3', 5'-di Cl		35.95 (35.78)	22	40	100	221	100	272
7.	2'-OH, 3', 5'-di I		55.10 (55.21)	33	30	20	32	100	144
8.	4'-OH		22.27 (22.66)	44	31	35	112	90	150
9.	4'-OH, 3',5'-di I Tetracycline Water-ethanol (90 : 10, v/v)		55.45 48 25	30 48 25	26	50	80	20	96
						95	305	105	350

The antibacterial and antifungal activity of all synthesised compounds were assessed following the standard methods<sup>17</sup> against two animal pathogens *Staphylococcus aureus*, *Escherichia coli* and two fungal phyto pathogens e.g., *Curvularia lunata* and *Helminthosporium oryzae*, respectively. For antibacterial activity, the filter paper discs were soaked in the solution of above compounds at a concentration of 150 ppm of compound in 10% ethanol and placed at the centre of the bacteria seeded agar plates (petridishes).

The petridishes were incubated at 26±1°C for 24h. The strength is reported in mm (Table 1) and results were standardised against tetracycline.

For antifungal activity, spore suspension was prepared from 5 day old PDA (Potato dextrose agar) slope cultures. Spore suspensions were placed in small petridishes. Solutions of different synthesised compounds were prepared in 90:10 (v/v) water : ethanol, and the concentrations of compounds (150 ppm) were adjusted in spore suspensions. Petridishes were incubated for 12 h under moist chambers. Aqueous ethanol (90:10, v/v) served as control. Percentage germination and germ tube length with effect of these compounds after a period of 12h was recorded by observing petridishes directly under microscope. These results are given in Table 1.

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## Vol. 71, Part I, 2001

### CONTENTS

#### Chemistry

Equalized electronegativity and chelation	<i>B.S. Sekhon</i>	...	1
Studies on synthesis of some novel bifunctional reactive dyes and their application on various fibres	<i>M.S. Patel, S.K. Patel and K.C. Patel</i>	...	5
Study of metal drug equilibria in solution	<i>Purushottam B. Chakrawarti, Mukta Chakrawarti and Pramila Maini</i>	...	13

#### Mathematics

Thermo-elastic stress distribution in three layered system	<i>Ashoke Das and Bimalendu Das</i>	...	21
Effect of rotation on thermal instability in Walters elastico-viscous fluid	<i>Pardeep Kumar</i>	...	33

#### Physics

Possibility of the simultaneous occurrence of potential minima along two crystallographic directions in an octahedral potential	<i>R.K. Tiwari and Prem N. Singh</i>	...	43
Liquid crystalline behaviour of alkoxybenzoic acids : A theoretical study	<i>S.N. Tiwari and Nitish K. Sanyal</i>	...	53
Dielectric study of pyridine-alcohol binary liquids at 25°C	<i>Ajay Chaudhari, Sunil Ahire, Milind Lokhande and Suresh Mehrotra</i>	...	75

## Vol. 71, Part II, 2001

### Chemistry

Synthesis and characterization of transition metal complexes of isatin oxime	<i>K. Laxmi, B. Sireesha and Ch. Sarala Devi</i>	...	85
Synergic extraction and trace determination of vanadium (V) as ternary chelate with cinnamohydroxamic acid and thiocyanate in mibk and its application	<i>A.K. Chakrabarti</i>	...	95
Synthesis and characterization of silatrane containing heterocyclic moieties : Part III	<i>Shailesh C. Verma, M. Nasim and P.S. Venkataramani</i>	...	105

### Mathematics

Stability of a stratified partially ionized plasma	<i>Aiyub Khan and P.K. Bhatia</i>	...	113
Boundary value problems of infinite plate weakened by a curvilinear hole	<i>Abdullah A. Badr</i>	...	125
Motions and affine motions in Finsler, Randers and Kropina Spaces	<i>U.P. Singh</i>	...	135
Free convection in MHD flow of a rotating viscous liquid in porous medium past a vertical porous plate	<i>N.P. Singh, Sarvesh Kumar Gupta and Atul Kumar Singh</i>	...	149
On the ultradistribution of Mellin transform	<i>P.K. Banerji and Deshna Loonker</i>	...	159

### Short Notes

Mixed ligand complex formation of $M^{2+}$ ions and quinoline and thymine	<i>Purushottam B. Chakrawarti and Ms. Sandhya Sharma</i>	...	173
A new spot test for the on field detection of dithiocarbamate fungicides	<i>H.S. Rathore, S. Mital, S. Kumar and Y.N. Singh</i>	...	177
Study of metal-drug equilibria in solution	<i>Purushottam B. Chakrawarti and Km. Sandhya Sharma</i>	...	185

## Vol. 71, Part III, 2001

### Chemistry

Synthesis of some new azo-coumarins and Schiffs bases as possible antibacterial agents <i>Sangeeta Bhatnagar, Kausik Ghosh and D.S. Seth</i>	... 189
On the coordinating behaviour of sulphamethoxazole towards some metal ions <i>B.S. Sekhon, Harvinder K. Sahai and H.S. Randhawa</i>	... 195
Synthesis and characterization of 8-hydroxy-quinoline-oxamide-formaldehyde terpolymers <i>Peter S. Lingala, L.J. Paliwal and H.D. Juneja</i>	... 205

### Mathematics

A uniform model for the storage utilization of <i>B</i> -tree-like structure <i>P.K. Mishra and C.K. Sharma</i>	... 213
Non-linear effects in the motion and stability of an inter-connected satellites system orbiting an oblate Earth <i>B.M. Singh, Ashutosh Narayan and R.B. Singh</i>	... 225
A common fixed point theorem in complete metric spaces by altering distances <i>K.P.R. Sastry and G.V.R. Babu</i>	... 237
Hall current effects on heat and mass transfer in flow of a viscous fluid <i>N.P. Singh</i>	... 243
Unsteady flow of visco-elastic Oldroyd fluid with transient pressure gradient through a rectangular channel <i>Shyamal Kumar Kundu and P.R. Sengupta</i>	... 253
Fluctuating free convective flow with radiation through porous medium having variable permeability <i>S.S. Tak and Arvind Maharshi</i>	... 263

### Physics

Study of photoconductivity in $(\text{B}_2\text{O}_3 - \text{ZnS})\text{Cu}$ (0.1%) Cl (1%) composite <i>Akhilesh Kumar, Sunil Kumar Srivastava and S.G. Prakash</i>	... 273
---	---------

### Short Note

Study of drinking water quality in Gorakhpur city <i>Alok Srivastava, Amitabh K. Srivastava, P.P. Srivastava and S.C. Prasad</i>	... 281
---	---------

## Author Index

Ahire, Sunil	75	Patel, K.C.	5
Babu, G.V.R.	237	Patel, M.S.	5
Badr, Abdullah A.	125	Patel, S.K.	5
Banerji, P.K.	159,321	Prakash, S.G.	273
Baseer, M.A.	351	Prasad, S.C.	281
Bhatia, P.K.	113	Randhawa, H.S.	195
Bhatnagar, Sangeeta	189	Rathore, H.S.	177
Chakrabarti, A.K.	95	Sahai, Harvinder K.	195
Chakrawarti, Mukta	13	Sanyal, G.S.	295
Chakrawarti, Purushottam B.	13,173,185	Sanyal, Nitish K.	53
Chaudhari, Ajay	75	Sastry, K.P.R.	237
Das, Ashoke	21	Sekhon, B.S.	1,195
Das, Bimalendu	21	Sengupta, P.R.	253
Devi, Ch. Sarala	85	Seth, D.S.	189
Ganguly, R.	295	Sharma, C.K.	213
Ghosh, Kausik	189	Sharma, Km. Sandhya	173,185
Gupta, Sarvesh Kumar	149	Shenan, Gamal M.	321
Juneja, H.D.	205	Singh, Atul Kumar	149
Khan, Aiyub	113	Singh, B.M.	225
Khurana, Sandhya	305	Singh, N.P.	149,243
Kumar, Akhilesh	273	Singh, Prem N.	43
Kumar, Pardeep	33	Singh, R.B.	225
Kumar, S.	177	Singh, Sarva Jit	305
Kundu, Shyamal Kumar	253	Singh, U.P.	135
Laxmi, K.	85	Singh, Y.N.	177
Lingala, Peter S.	205	Sireesha, B.	85
Lokhande, Milind	75	Sivaramakrishna, Akella	289
Loonker, Deshna	159	Srivastava, Alok	281
Maharshi, Arvind	263	Srivastava, Amitabh K.	281
Maini, Pramila	13	Srivastava, P.P.	281
Mehrotra, Suresh	75	Srivastava, Sunil Kumar	273
Mishra, P.K.	213	Subbanwad, G.R.	351
Mital, S.	177	Tak, S.S.	263
Narayan, Ashutosh	225	Tiwari, R.K.	43
Nasim, M.	105	Tiwari, S.N.	53
Nath, P.K.	295	Venkataramani, P.S.	105
Paliwal, L.J.	205	Verma, Shailesh C.	105
Pande, L.K.	339	Vidhute, Y.B.	351

# Subject Index

4,4'-methylene-bis ortho anisidine	5	Hall current	243
8 HQQF	205	Hane-transform	21
Alcohols	75	Heat flux	21
Alkoxybenzoic acids	53	Heat source	243
Antimicrobial agents	351	Heterocyclic substituted silatranes	105
Axisymmetric	21	Hilbert space	237
Bis-azo reactive dyes	5	Hydromagnetic stability	113
B-trees	213	IR	5,351
CNDO	53	Inter connected satellites	225
Cation-exchange resin	177	Ionic crystals	43
Cd(II)	13	Isatin oxime	85
Cephalexin complexes	13	Kropina space	135
Chalkones	351	Layered problem	21
Chelation	1	Linear temperature distribution	21
Chemistry	1,5,13,85,95,105, 173,177,185,189,195, 205,281,289,295,351	Liquid crystals	53
Common fixed points	237	M2+-quinoline-tymine equilibria	173
Complete matric space	237	Magnetic field	149
Composites	273	Magnetic & spectral studies	85
Convex and starlike convex functions	321	Mass transfer	243
Coordinating behaviour	195	Mathematics	21,33,113,125,135, 149,159,213,225,237, 243,253,263,305,321
Copper-ammonia complex	177	Mellin transform	159
Coumarin derivatives	189	Mesogen	53
Dielectric parameters	75	Metal complexes	295
Dithiocarbamates	177	Metal ions	195
Earth oblateness	225	Metal-drug equilibria	13,185
Elasto-static	21	Mg(II) complexes	13
Electron proton pairing	339	Mixed-ligand complexes	13
Electronegativity	1	Molecular interactions	53
Elemental analysis	351	Monoclinic media	305
Ferricyanide	289	Non-linear oscillation	225
Finite resistivity	113	N-CHA	95
Finite temperature theory	339	N-(2-tri fluoromethyl)	189
Finsler geometry	135	Octahedral potential	43
Fractional differential and integral operators	321	PMR	351
Fungicides	177	Phenyl malonamic acid	189
Generalized visco-elastic fluid	253	Photoconductivity	273
		Physico-chemical & bacteriological properties	281
		Physics	43,53,75,273,339

Polarization	43	Tercopolymer	205
Porous medium	149	Ternary chelate	95
Pyrazine carboxylic acid hydrazides	295	Ternary complexes	173,185
Pyridine	75	Testing function space	159
Randers space	135	Thermal instability	33
Rectangular channel	253	Time domain reflectometry	75
Reflection	305	Titrimetry	289
Riordan array	213	Transesterification	105
Rotating viscous liquid	149	Transient pressure gradient	253
SCN	95	Transition metal complexes	85
Schiffs bases	189,295	Transmission	305
Selfmaps	237	UO <sub>2</sub> (VI)	295
Semi-conductors	273	Ultradistribution	159
Sherwood number	243	Uniform rotation	33
Silk	5	Univalent and p-valent functions	321
Soret number	243	Unsteady flow	253
Spectral studies	189	VO(IV)	295
Stability constant	13	Vanadium	95
Stability of bivalent metal complexes	173	Viscose rayon	5
Stability of metal chelates	1	Viscosity	113
Stability of quinoline complexes	173	Visco-elastic Oldroyd fluid	253
Stratified plasma	113	Walters elastico-viscous fluid	
Sulphamethoxazole	195	(model B')	33
Sulphide sulphur	289	Water quality	281
Synergic extraction	95	Waves	305
		Wool	5
		X-ray astronomy	339

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